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The three preceding issues of this Journal are dated March 1961, May 1961, and July 1961.

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EFFECTS OF HYDROGEN SULFIDE ON CONCRETE STRUCTURES<sup>a</sup>

By Bernal H. Swab, F. ASCE<sup>1</sup>

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SYNOPSIS

Disintegration of concrete structures by acids formed from hydrogen sulfide prevails in many areas. In other areas with seemingly similar conditions no disintegration occurs. Many factors must be considered in the design of sewers in which this problem might occur. More research is needed to determine all the causes and the reasons why disintegration does not occur in all localities under similar conditions.

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INTRODUCTION

Over the years this subject has received considerable publicity, some fair and some unfair. This paper will attempt to clarify some of the conflicting points and will examine some of the questionable areas. Theory will be held to a minimum because Richard Pomeroy, F. ASCE, and other capable engineers have covered this field and will continue to present improvements on the theoretical phase of this problem.

No statements herein are intended to promote or condemn any individual product, but all engineers and municipal maintenance forces must be made aware of the serious problems involved in hydrogen sulfide corrosion. Some statements will repeat already published information to emphasize important points.

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Note.—Discussion open until February 1, 1962. To extend the closing date one month, a written request must be filed with the Executive Secretary, ASCE. This paper is part of the copyrighted Journal of the Sanitary Engineering Division, Proceedings of the American Society of Civil Engineers, Vol. 87, No. SA 5, September, 1961.

<sup>a</sup> Presented at the April, 1961 ASCE Meeting in Phoenix, Ariz.

<sup>1</sup> San. Engr., Camp, Dresser and McKee, Boston, Mass.

The term, "concrete and concrete structures," as used herein, is meant to include all pipe and other structures containing portland cement.

### HYDROGEN SULFIDE PROBLEM

The general statement has been made that hydrogen sulfide has no place in any sewer system, regardless of the materials used in construction, for the following reasons:

1. The typical rotten-egg odor emerged from manholes, causes a nuisance, and lowers property values.
2. Hydrogen sulfide is an indicator of septic sewage. This type of sewage is more difficult to treat than fresh.
3. The gas attacks paint on sewer structures as well as that on other neighborhood structures.
4. The gas is toxic. As concentration increases, it impairs the sense of smell. Far too many sewer maintenance workers have lost their lives because of this gas.
5. The explosive range of hydrogen sulfide is from 4.3% to 46.0% by volume in air.<sup>2</sup>
6. In the form of sulfuric acid, it attacks structures containing portland cement as well as many metals.

No corrosion will occur where sewers flow full. The attack will occur above the water line as shown on Fig. 1. As the attack continues, the pipe may disintegrate to the extent that failure occurs, as shown on Fig. 2. Ultimate failure can occur to reinforced pipe as indicated on Fig. 3. When failure occurs, cave-in of streets may occur as shown in Fig. 4.

Corrosion from hydrogen sulfide should not be confused with corrosion caused by highly acid trade wastes. Typical acid-waste corrosion of the invert is shown on Figs. 5 and 6. Hydrogen sulfide corrosion will occur in the upper portion of the pipe, above the water level, as shown on the previous figures.

*Distribution of Corrosion Problem.*—Some authorities have stated that this corrosion problem is confined to Southern California, but research within the scope of this paper indicates the problem in general exists in certain areas of Southern California, Arizona, New Mexico, Texas, Gulf Coast Area, the Middle Atlantic Coastal States, Iowa, Nebraska, South Dakota, North Dakota, Kansas, Missouri, Oklahoma, Hawaii, South Africa, and Australia.

There are known isolated cases in many other areas.

*History.*—In 1881 Rudolph Hering<sup>3</sup> gave the first indication of possible trouble to concrete structures from hydrogen sulfide. This article included statements that are applicable today.

Hering examined design, ventilation and maintenance among other phases. A review of this paper will reveal that we have made little progress in the design of sewers since that time. Hering listed the factors involved in the intensity of the odor and advocated simplicity of design and high degree of maintenance.

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<sup>2</sup> "Safety in Wastewater Works," FSIWA Manual of Practice No. 1, 1959, p. 17.

<sup>3</sup> "Sewerage Systems," by Rudolph Hering, *Transactions*, ASCE Vol. X, 1881, p. 366.



FIG. 1.—CORROSION OF SEWER IN MONROE, LA.



FIG. 2.—TOTAL COLLAPSE OF SEWER PIPE IN PHILADELPHIA, PA.



FIG. 3.—FAILURE OF 48-IN. REINFORCED CONCRETE PIPE



FIG. 4.—CAVE-IN OF STREET FROM COLLAPSED  
SEWER IN MOBILE, ALA.

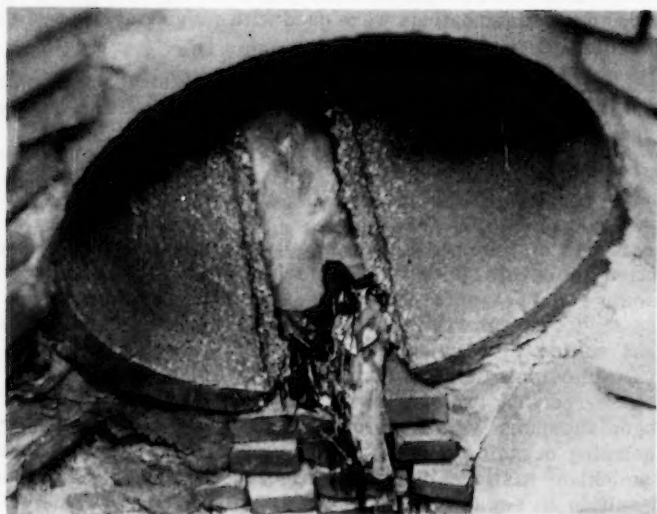


FIG. 5.—ACID CORROSION OF CONCRETE PIPE IN ATLANTA, GA.

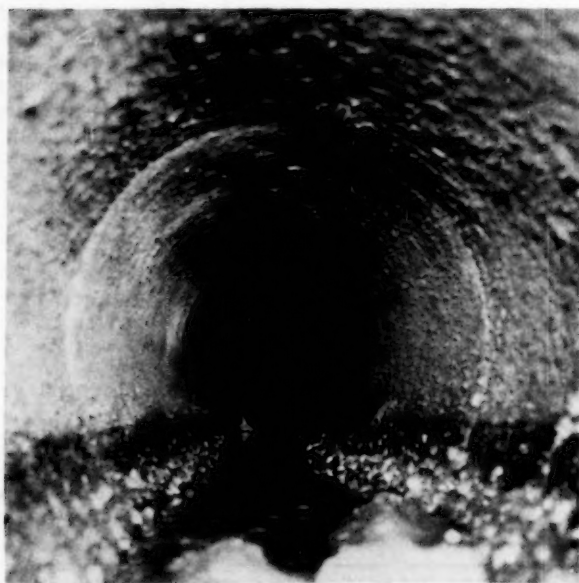


FIG. 6.—CORROSION OF CONCRETE PIPE BY HIGH CONCENTRATION OF ACID WASTES IN SEWER IN HICKORY, N. C.

The protection of concrete structures has been recognized for many years. In the 1920's, protective coatings were used with varying degrees of success. In 1925, Karl Imhoff wrote:

"I am glad to confirm that we have used in our Ruhrverband in Essen the paint, Inertol, with good success, especially in the five-mile concrete sewer for protecting concrete against sewer gases."

### CONTRIBUTING FACTORS

The corrosion of concrete in sewerage structures results from the metabolic activity of several species of bacteria. Most of the bacteria and fungi found in sewage tend to oxidize sulfur and sulfur containing compounds. However, there are a few that will reduce sulfur and sulfur-containing compounds to hydrogen sulfide. The most important sulfur-reducing bacteria belong to the genus *Desulfovibrio* that will reduce sulfates to hydrogen sulfide.

The sulfate-reducing bacteria are strictly anaerobic and use the sulfate as the hydrogen acceptor. Various organic materials can be used as hydrogen donors including organic acids, alcohols, amino acids, and carbohydrates.<sup>4</sup>

H. Heukelekian<sup>5</sup> has investigated the effect of nitrates on the production of hydrogen sulfide in sewage. He concludes that hydrogen sulfide will not be produced in sewage until the bacteria have first utilized all of the dissolved oxygen and have reduced all of the nitrates. When no further oxygen is available from dissolved oxygen or nitrates, the bacteria present in the sewage will begin to reduce the sulfates and form hydrogen sulfide.

Hydrogen sulfide is a highly reduced state for sulfur, and it is readily oxidized by dissolved oxygen or by many species of bacteria when oxygen is available. In a sewer, the necessary oxygen may be obtained by aeration or chlorination of the sewage and oxygen found dissolved in the condensate at the crown of the sewer. In the condensate at the crown of the sewer, the hydrogen sulfide frequently is oxidized to sulfuric acid. In the sewage itself, competing reactions usually result in the oxidation of the hydrogen sulfide to sulfates when dissolved oxygen is present.

Some of the known physical and chemical conditions that contribute to bacterial production of hydrogen sulfide are listed as follows:

1. High BOD
2. High sulfates in domestic sewage and industrial wastes
3. High sewage temperatures
4. Sluggish and stagnant flow conditions

In localities in which all conditions are favorable for bacterial growth, corrosion does not occur uniformly or predictably. Some unknown factors seem to determine that one location will be attacked, whereas another with similar conditions will remain unaffected. Some structures have been immune from attack over a period of years and suddenly begin to corrode. This may be due to a change in the biological balance of the bacteria in the sewage resulting in

<sup>4</sup> "Transformations of Sulfur by Microorganisms," by Robert L. Starkey, *Industrial and Engineering Chemistry*, Vol. 48, 1956, p. 1429.

<sup>5</sup> "Effect of the Addition of Sodium Nitrate on Hydrogen Sulfide Production and B.O.D. Deduction," by H. Heukelekian, *Sewage Works Journal*, Vol. 15, 1943, p. 225.

rapid generation of sulphuric acid and corrosion of concrete causing a change in the flow characteristics.

High concentrations of sulfates and nitrates in the water supplies and their relation to hydrogen sulfide corrosion were reviewed. Water supplies of over 1,300 communities<sup>6</sup> were compiled and high values plotted in accordance with their concentrations as shown on Figs. 7 and 8.

These indicate that high sulfates are present in areas in which corrosion is not prevalent. In some areas, high nitrates may retard bacterial production of hydrogen sulfide, but they certainly do not do so in the known areas of corrosion. This may be due to an imbalance of organisms and the absence of some of the chemical and physical contributors.

### PREVENTION OF HYDROGEN SULFIDE BUILD-UP

Methods of preventing hydrogen sulfide build-up with resultant corrosion of concrete structures have been published. Some of these are applicable to new design and others to maintenance. Each will be considered separately.

### DESIGN OF NEW STRUCTURES

In designing new systems, the engineer must consider all contributing factors and complete a design that is economical for the client to construct and maintain. Such economical designs must consider topography, grades of sewers, ventilation, pumping stations, type of wastes, disposal of wastes, materials of construction, and operating and maintenance costs.

The Pomeroy-Bowlus Theory<sup>7</sup> is very useful in designing sewers wherein there will be the least probability of sulfide build-up. Revisions are being made to cover some of the unknown factors. Nevertheless, the engineer should be cautioned regarding proper assumptions in applying this or any other formula that may be proposed.

**Peak BOD.**—In so many cases, design computations consider daily average BOD and temperatures. Daily peak BOD may run as much as 50% to 100% over the average and should be considered.

**Prediction of Future BOD.**—Accurate prediction of future BOD levels is very difficult because the engineer cannot predict what type of industries may be built and type of wastes that will be discharged into the sewers. Many industries will increase flows by only a small amount, but their load may increase the BOD by many times that which was anticipated.

Clyde L. Palmer, F. ASCE and Isadore Nusbaum, M. ASCE<sup>8</sup> report, on the basis of study made in Detroit, Mich. that if 100% of a community installed garbage grinders, the BOD of the sewage from that community would be increased 20% to 25%.

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<sup>6</sup> "The Industrial Utility of Public Water Supplies in the United States," U. S. Geol. Survey Water Supply Papers, Nos. 1299 and 1300, 1952.

<sup>7</sup> "Progress Report on Sulfide Research," by Richard Pomeroy and Fred Bowlus, *Sewage Works Journal*, Vol. 18, 1946, p. 597.

<sup>8</sup> "Ground Garbage Studies in Detroit," by Clyde L. Palmer and Isadore Nusbaum, *Sewage and Industrial Wastes*, Vol. 23, 1946, p. 1113.



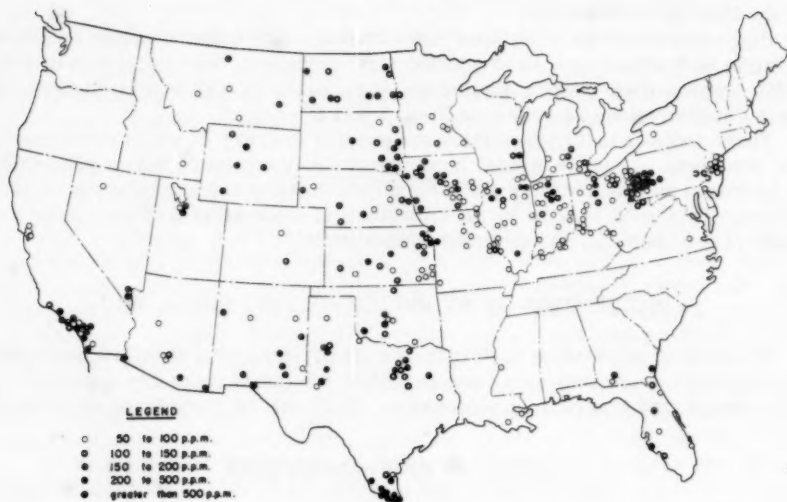


FIG. 7.—SULFATE CONCENTRATIONS IN PUBLIC WATER SUPPLIES OF THE UNITED STATES

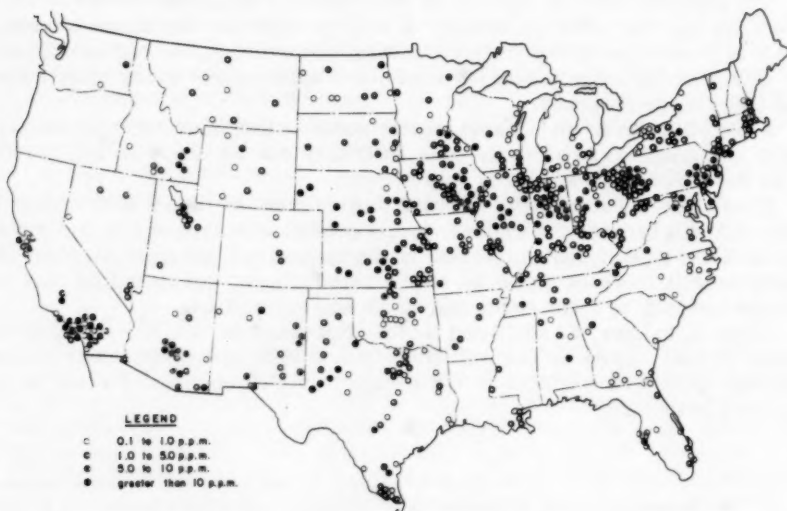


FIG. 8.—NITRATE CONCENTRATIONS IN PUBLIC WATER SUPPLIES OF THE UNITED STATES



It may seem inconsistent, but in some locations large increases in BOD loading have not increased hydrogen sulfide build-up. Answers to such apparent inconsistencies require additional research.

**Prediction of Future Temperatures.**—Future temperature prediction is as difficult as that of BOD, because a new contributor can increase temperatures far above normal sewer temperatures and cause trouble. Increase in use of household appliances using hot water can increase temperatures above those now considered normal. Some temperature measurements indicate that maximum sewage temperatures from residences now vary from 100°F to 136°F.

**Prediction of Velocities in Sewers.**—Some controlling agencies set up minimum design velocities to be used in the design of sewer systems. This minimum is normally 2 fps. Some systems have lower velocities and evidently give no trouble, whereas others designed for this minimum or even greater velocities have caused maintenance problems.

Present (1961) minimum velocities should be reconsidered because with the increased use of garbage grinders such items as egg shells, ground bones, and broken glass will tend to settle out and increase maintenance problems. George W. Reid, M. ASCE and Tien-Sheng Yang, A.M. ASCE<sup>9</sup> reported that low velocities in sewers permit the more rapid build-up of slimes on the barrel of the sewer that further decrease the velocities. Their studies with partially filled pipes at the University of Oklahoma, Norman, Okla. indicated that in 12 weeks the original velocity of from 1.38 fps in a clean sewer might be decreased as much as 35%, whereas with original velocities of from 1.59 fps to 1.70 fps, the decrease in velocity was only about 12%. Approximately 12% by weight, more slimes accumulated on the sewer barrels at the lower than at the higher velocities. The decrease in velocity and the amount of slimes accumulated varied with different types of pipe.

Additional research on effect of slimes on friction factors and velocities and transporting velocities of partially full pipes would help clear up many questions.

**Force Mains.**—In designing new systems, the engineer is often faced with practical design of lift stations and force mains. Even if his crystal ball anticipates future flows and BOD, he is faced with the difference between present and future conditions. In most cases, there will be a very wide range between present minimum and future maximum flows. Generally, pumps can be designed to cover any anticipated range, but sizing of wet wells and force mains becomes the problem.

If wet wells are made large enough for future conditions, long periods of storage may exist during present low flows and contribute to septicity and ultimate hydrogen sulfide build-up. Ample ventilation should always be provided to such wells to promote dry walls and carry away any hydrogen sulfide gas that may be generated. Failure to do so might produce conditions as shown on Figs. 9 and 10. Wet wells constructed with temporary walls, or in sections, can be used to reduce the detention period during low flow.

Long detention periods in force mains during low flows can generate hydrogen sulfide. The force main will be full and no corrosion will occur; but when discharged into partially full gravity sewers and wet wells, the hydrogen sulfide generated in the main will be released and in most cases cause corrosion.

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<sup>9</sup> "Sewer Slimes Cause Odors," by George W. Reid and Tien-Sheng Yang, *Waste Engineering*, September, 1959.



FIG. 9.—CORRODED CONCRETE IN WET WELL IN  
CLOVIS, N. M.



FIG. 10.—CORRODED CONCRETE IN WET WELL IN  
HAMPTON ROADS, VA.

An example of this is shown on Fig. 11. An 8 in. force main approximately 3,500 ft long had a maximum retention period at low flow of 18 hr. The force main discharged the sewage into a 10 in. concrete gravity sewer, 15,000 ft long, flowing at a velocity of about 2 fps when the pumps were operating. It is quite possible that the sulfide build-up increased in this section of the line. The grade of the end section of the sewer was steeper than the 15,000 ft section, and flowed partially full at about 2 fps. This portion of the sewer, and the adjacent 10 ft of the flatter grade section, disintegrated.

Pomeroy has written a number of articles covering the interjection of air into force mains to prevent hydrogen sulfide build-up in these mains. In one

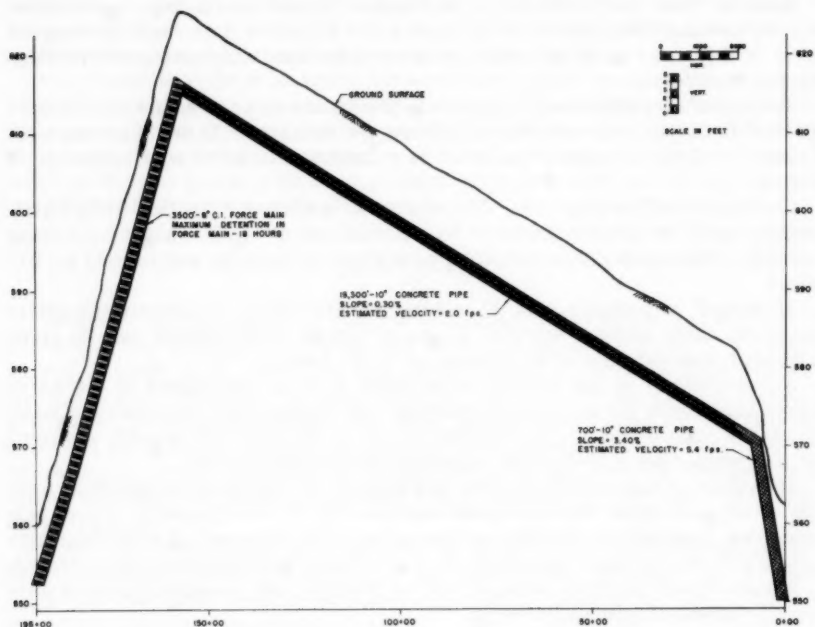


FIG. 11.—SULFIDE BUILD-UP IN FORCE MAIN AND GRAVITY SEWERS IN FORT WORTH, TEX.

article,<sup>10</sup> the technique of sulfide control by air injection is covered in detail and should be considered in the design of force mains with long detention periods. His studies indicate that in most cases air injection will prevent hydrogen sulfide build-up and in any case will greatly reduce generation.

**Reduction of Turbulence in Flow.**—After hydrogen sulfide has built up in force mains or in flowing-full sewers, the gas will be released more rapidly where turbulence exists. Such turbulence can exist at the outlet end of the

10 "Generation and Control of Sulfide in Filled Pipes," by Richard Pomeroy, *Sewage and Industrial Wastes*, Vol. 31, 1959, p. 1082.

force mains with free fall, at sudden changes to steeper grades in sewers, at sharp bends in sewer lines flowing partially full, and within manholes.

Turbulence might be reduced by having submerged discharge of force main and reducing sudden changes in grade and direction of sewers. In some instances, this might be more costly than using materials that will withstand the action of hydrogen sulfide.

**Ventilation.**—Ample ventilation through sewers and wet wells will tend to carry away the generated hydrogen sulfide, supply additional oxygen to the sewage and keep the walls free of moisture and reduce the tendency for sulfuric acid attack. Ventilation is particularly important in locations of turbulent flow.

Some of this ventilation might be obtained by proper venting of manholes. In many communities, the manhole covers are solid and the crack between the cover and frame sealed with various materials, thus eliminating any ventilation through covers.

Where forced ventilation is required, provisions must be made for the disposal of the rotten-egg odor from causing any nuisance. In some areas, such methods will prove costly and must be considered from an economical point of view.

**Control of Sulfide Sources.**—The amount of wastes contribution to  $H_2S$  generation could be greatly reduced by elimination of trade wastes containing sulfides, reduction of concentration of sulfates in sewage, and partial purification.

Municipal ordinances should prohibit the discharge of any wastes detrimental to their sewage system. In most cases, these wastes can be given sufficient pretreatment to be acceptable to the system.

Any reduction in the amount of sulfates in incoming wastes or the water supply may lower the sulfate concentration to a safe level. However, in many cases, the sulfate content in domestic sewage is sufficient to provide a source for the production of damaging quantities of hydrogen sulfide.

**Materials of Construction.**—The engineer must design an economical system using materials that are most suitable for the conditions to be met. On occasions, pressure is brought on the engineer to allow use of substitute materials inferior to those specified. It is gratifying that this practice is fading.

Joints should be tight and of a type to prevent root penetration which ultimately causes clogging of sewers.

If hydrogen sulfide build-up cannot be prevented, corrosion-resistant materials must be specified. In sewers of smaller sizes, clay or other acid-resistant pipe can be used, but in larger sizes concrete products must be protected by an impervious coating or by additional thickness of sacrificial concrete.

Use of sacrificial concrete to combat corrosion is questionable, because neither the rate of corrosion, nor the depth of penetration can be predicted with any degree of certainty. No type of pervious lining will provide satisfactory protection against hydrogen sulfide corrosion. The most minute hole in the wall or joint of the lining will permit the passage of acid into the concrete and eventually cause corrosion. The pipe should be as impervious as possible to prevent ground water from percolating through the pipe walls and forcing the lining away from the walls. The required tests on all pipe might be revised to include a hydrostatic pressure test similar to that used in testing clay pipe on the west coast of the United States.

Pipe joints covered with liners should be as watertight as the barrel of the pipe. Additional research should be made to accomplish this aim.

Clay liner plates with cement joints in concrete pipe have proven unsatisfactory in many instances. The joints disintegrate and the pervious liner plates permit the acid to pass through the plate with ultimate corrosion of the concrete. When placed with cement or other rigid joints, there will be a tendency for the plate or cement joint to crack when the backfill load is placed on the pipe.

Some authorities recommend the use of limestone aggregate in concrete as a retardant to corrosion. This statement should be accepted with caution, because the compressive strength, specific gravity, abrasion resistance, soundness, absorption, and other qualities of limestone can vary over a wide range in the same quarry. New tests might be developed to determine the qualities that best resist corrosion from hydrogen sulfide.

No matter how large or small the sewerage project may be, the materials should be installed in the best possible manner. Negligence on the part of the engineer to require proper bedding and backfill to prevent possible movement of the structure and to require proper cleaning of ends of pipes and gaskets will result in an inferior finished product. The owner should receive the best possible system for the money expended.

## MAINTENANCE

*General.*—During low flows and low velocities, heavy solids and silt tend to deposit in the bottom of sewers, and slimes form on the sides of the sewer barrel. Unless peak velocities are large enough to flush out these restrictions, pooling of sewage will provide ideal locations for the growth of hydrogen sulfide producing bacteria. As most maintenance forces know, manholes are used as dumping receptacles for considerable volumes of refuse. On one occasion, the writer observed the clogging of a 36-in. sewer by orange crates and on another, by a dead cow dumped into a manhole in front of a horse meat canning plant.

Sewer blockage from these and similar cases require removal to prevent odors, damage to the surrounding property, and eventual damage to the sewers. Care must be taken in the selection of cleaning methods to prevent damage to any lining used for protection.

In the upper reaches of sewers, minimum size pipes are used that are oversize for the present contributing population, and in other cases, sewers are designed to provide additional capacity for future extension. In such locations, slime growths and low velocity depositions will be encountered and maintenance required. This can be accomplished by a regular flushing program. The flushing can be facilitated by installing accessible terminal clean-out structures that can be easily removed if future extension of the sewer is required. In old systems, this problem was partially overcome by installing flush tanks. In many locations, these structures were neglected and failed to function. Some are operating today and adequately flushing the sewers to prevent deposits from accumulating.

It is difficult in some areas to have the governing bodies provide ample funds for proper sewer maintenance in their budgets because the sewers are underground; nobody can see them and few realize their importance. Com-

munities should realize their large investment in these unseen structures, as well as the additional financial burdens that could be placed on them by improper maintenance.

*Prevention of Hydrogen Sulfide Build-Up.*—A number of methods to be used to prevent hydrogen sulfide build-up in existing systems have been published. Some have been considered under design.

The following may be listed under operation and maintenance:

*Elimination of Trade Wastes Containing Sulfides.*—Ordinances should prohibit dumping of wastes that are harmful to the sewerage system.

*Partial Purification of Sewage.*—This method in existing systems may require a multiplicity of treatment units that would prove very costly. New industries should be required to pretreat any objectionable wastes before discharge into sewers.

*Aeration of Force Mains.*—On existing force mains, this may provide a means of preventing build-up providing the mains do not contain a number of pockets to accumulate the air. Multiple compressor stations might be installed with air release valves installed on all high points on the line.

*Dosage with Chemicals.*—Many different chemicals can be used to deter the formation of hydrogen sulfide, but all are costly. It is quite possible that the administration that installed the sewers will provide funds because they are acquainted with the problem. As members of the administration change and new administrators follow through on the universal campaign promise of cutting taxes, they are prone to cut the budget item for chemicals used in sewer maintenance because the electorate cannot see the results of eliminating any required chemical treatment until full collapse of the sewers occur.

*Running Sewers Full.*—Such a program to prevent corrosion may only cause the transfer of the trouble to another location. While flowing full, additional hydrogen sulfide may be generated and increase the damage unless manholes are well vented. If the sewers were flowing full at minimum flows, there would be no capacity available for average or peak flows, unless a program was inaugurated to add varying quantities of dilution water with varying flows. This might be very costly.

*Venting Sewer Connections.*—Building ordinances or plumbing codes sometimes require that a vent be placed on the street side of the first trap into a building. This vent acts as a chimney in drawing moisture vapor and sulfide-laden air from common sewers and house connections before damage can be done to the pipe or joints.

## SUMMARY

The problem of hydrogen sulfide corrosion has been recognized over a number of years, but little has been done about it. One special interest group concentrated on condemning the product of another while the other group publicized the years their product had been in use with little reference to the corrosion problem.

Failures of structures make it imperative that a solution to this problem be obtained. Research by Pomeroy and other capable investigators have made considerable progress, but additional research is necessary to clarify the action of some unknown quantities contributing to this problem.



This problem is not localized. This corrosion has occurred over widely distributed areas in the United States. Under proper conditions for bacterial growth, it can happen in any system.

All contributing factors should be thoroughly considered in the design of sewerage systems. The more important of these may be listed as follows:

1. High temperature of sewage
2. High BOD
3. Low velocity of flow
4. Detention period in force mains and wet wells
5. Degree of turbulence in partially filled conduits
6. Lack of ventilation
7. Use of corrosion-resistant materials
8. Protective coatings

Improvements can be made in many materials to make them more corrosion resistant. Progress is required in the development of an economical acid-resisting cement and better criteria for the determination of the most acid-resisting aggregates.

Specifications can be improved for tests on porosity of pipe to obtain more impervious pipe.

Continued improvement should be made on joint design and materials for several types of pipe. Further development of plastic pipe for sewers may accelerate such improvements. Some types of plastic linings for sewers and other structures have proved satisfactory to date, but additional years of service will be needed to determine the life of these materials. Some sprayed and brush coatings have been found lacking in their ability to resist hydrogen sulfide corrosion. Probably this is due to incomplete surface coverage or some type of abrasion removing the coating. More research is needed to produce efficient and economical linings.

An adequate maintenance and cleaning program must be sold to the community fathers as well as to the maintenance forces of municipalities so that all will understand the economic and sanitary importance of good maintenance.

The first of these is the fact that the human mind is not a blank slate, but is filled with a vast amount of knowledge and experience which is inherited from our ancestors. This knowledge is not only passed on to us by our parents, but also by our teachers and by the society in which we live. It is this knowledge which enables us to learn from our mistakes and to improve ourselves and our race.

The second of these is the fact that the human mind is not a passive receiver of knowledge, but is an active seeker of it. We are constantly asking questions and trying to find answers. We are constantly learning from our experiences and from the experiences of others. It is this active seeking of knowledge which enables us to progress and to improve ourselves and our race.

The third of these is the fact that the human mind is not a single entity, but is composed of many different parts. There is the intellect, which is the part of the mind that is responsible for reasoning and logic. There is the emotions, which are the part of the mind that is responsible for feelings and passions. There is the will, which is the part of the mind that is responsible for decisions and actions. It is the interaction of these different parts of the mind which enables us to think, feel, and act.

The fourth of these is the fact that the human mind is not a static entity, but is constantly changing. We are constantly learning new things and forgetting old things. We are constantly growing and developing. It is this constant change which enables us to adapt to our environment and to improve ourselves and our race.

The fifth of these is the fact that the human mind is not a solitary entity, but is a social entity. We are constantly interacting with others and learning from them. We are constantly sharing our knowledge and experiences with others. It is this social interaction which enables us to progress and to improve ourselves and our race.

The sixth of these is the fact that the human mind is not a purely rational entity, but is a purely emotional entity. We are constantly driven by our emotions and our passions. It is these emotions and passions which enable us to love, to hate, to hope, and to despair. It is these emotions and passions which enable us to live and to feel.

The seventh of these is the fact that the human mind is not a purely individual entity, but is a purely collective entity. We are constantly influenced by the society in which we live. We are constantly learning from the customs and traditions of our society. It is this collective influence which enables us to progress and to improve ourselves and our race.



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ADHESION IN FILTERS

By Vladimir Mackrle<sup>1</sup> and Svatopluk Mackrle<sup>2</sup>

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SYNOPSIS

By consideration of the van der Waals and hydrodynamic forces acting in a filter, it is possible to formulate a dimensionless group, known as the adhesion criterion, that includes parameters affecting the removal of suspended particles by the filter. This adhesion criterion is related to the Reynolds number in a manner independent of the nature of the suspended material and of the filter media.

With knowledge of the hydrodynamic characteristics of the filter, one can determine the concentration-depth curve characteristic. With given empirical information for aluminium and ferric hydroxide flocs, the reduction in suspended solids concentration can be calculated for a filter of any media grading, of any given depth.

Development of the form of the adhesion criterion and filtration equations dealing with the reduction in concentration of non-homogeneous suspensions in non-homogeneous (graded) filters is described.

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ANALYSIS OF THE ADHESION FORCES

*Notation.*—The letter symbols adopted for use in this paper are defined where they first appear, in the illustrations or in the text, and are arranged alphabetically, for convenience of reference, in the Appendix.

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*Note.*—Discussion open until February 1, 1962. To extend the closing date one month, a written request must be filed with the Executive Secretary, ASCE. This paper is part of the copyrighted Journal of the Sanitary Engineering Division, Proceedings of the American Society of Civil Engineers, Vol. 87, No. SA 5, September, 1961.

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Of interest in the field of filtration is the nature of the adhesion forces between the suspended particles and the filter grains in an aqueous medium.

It has been shown that the interactive force between a particle and an infinite surface is given by

$$F_i = \frac{A D_i}{12 r^2} \dots \dots \dots (1)$$

in which A is the constant of van der Waals attraction,  $D_i$  denotes the diameter of the particle and r refers to the distance between the surface of the particle and the infinite surface. Eq. 1 holds for a limiting distance of 100 Angstrom units ( $10^{-2}$  microns).

### FORMULATION OF THE FILTRATION EQUATION

Assuming that curvature of a filter grain can be neglected compared with the size of a floc particle, Eq. 1 can be used in which  $D_i$  is the floc particle diameter.

The hydrodynamic force acting on a particle in a streaming fluid is given by the Stokes equation

$$F_h = 3 \pi \mu D_i v \dots \dots \dots (2)$$

in which  $\mu$  is the viscosity of the fluid and, v denotes the fluid velocity relative to the particle.

In equilibrium the force attracting a particle to the surface of a filter grain is balanced by the hydrodynamic force, if the force vectors have the same line of action.

$$3 \pi \mu v_r = \frac{A}{12 r^2} \dots \dots \dots (3)$$

in which  $v_r$  is the particle velocity normal to the surfaces of the particle and filter grain.

The Navier-Stokes differential equation expresses the component of the velocity in the required coordinate direction. By the use of similitude criteria it can be shown that the dimensionless group  $\frac{A}{v_r \mu r^2}$  is constant for a given set

of filtration conditions and is given the symbol Ma.

The hydrodynamic situation in the filter can be completely expressed by the following dimensionless groups: Froudes number, Euler number, Ho number ( $= \frac{vt}{1}$ , expressing velocity conditions), Reynolds number, and adhesion criterion in the form

$$f(Ma, H, F, E, R) = 0 \dots \dots \dots (4)$$

In practice with the filter under pressure, isothermal conditions with steady state flow it follows that

$$Ma = f(R) \dots \dots \dots (5)$$

The Reynolds number contains a length term, specified by the Carman-Kozeny expression for hydraulic radius:

$$\text{hydraulic radius} = \frac{\text{porosity}}{\text{specific area of grains}} = \frac{w}{p} \dots \dots \dots (6)$$

The velocity term in  $R$  is the interstitial velocity given by

$$\frac{\text{approach velocity}}{\text{porosity}} = \frac{v}{w} \dots\dots\dots (7)$$

This yields

$$\frac{A}{v_r \mu r^2} = f \left( \frac{\rho v}{\mu p} \right) \dots\dots\dots (8)$$

in which  $\rho$  is the density of the fluid.

#### CONSIDERATION OF THE VAN DER WAALS CONSTANT

By quantum-mechanical calculations on the basis of work of various authors, it is possible to obtain the value of  $A$  between various materials in an aqueous solution. Such a list of values is given in Table 1, in which it can be considered that  $\text{Fe}(\text{OH})_3$  or  $\text{Al}(\text{OH})_3$  compact floc is the material to be filtered through a bed of either crystal silica (sand or glass spheres),  $\text{CaCO}_3$  granules, metallic lead spheres, or carbon grains.

TABLE 1.—CALCULATED VALUES OF THE CONSTANT  $A \times 10^{-12}$  FOR THE INTERACTION OF COMPACT SOLID PARTICLES IN AN AQUEOUS MEDIUM

Attracted material	Attracting material in the aqueous medium					
	$\text{Al}(\text{OH})_3$	$\text{Fe}(\text{OH})_3$	$\text{SiO}_2$ crystal	$\text{CaCO}_3$	Pb metal	C graphitic
$\text{Al}(\text{OH})_3$	1.26	1.42-1.45	1.27	1.40	1.27	1.30
$\text{Fe}(\text{OH})_3$	1.42-1.45	1.77-2.0	1.41-1.45	1.68-1.76	1.71-1.85	1.70-1.77

The steps in the calculation of  $A$  require knowledge of the molecular weight, number of valency electrons per molecule, the refractive index, the dipole moment, and the density of various materials involved, including water. These values are obtainable from international critical tables. For calculating the resulting  $A$  in aqueous solution, Hamaker-Verwey-Overbeek's equation must be used:

$$A = \frac{A''}{n_0^2} = \frac{1}{n_0^2} (A'_{12} + A'_{00} - A'_{10} - A'_{20}) \dots\dots\dots (9)$$

in which  $A'$  values are the Hamaker-van der Waals' constant for interaction of various materials in a vacuum, subscripts 1, 2, 0 indicate these interacting materials (0 for water), and  $N_0$  is the refractive index of pure water.

Values in Table 1 are calculated for compact materials. When interacting particles contain water inside, as is the case for many suspensions, their interaction constant  $\bar{A}$  may be calculated from the  $A$  value for compact particles as follows: designating the volume fraction of dense material in particle 1 by  $u_1$ , similarly in 2 by  $u_2$  [so that the water volume fraction in 1 is  $(1 - u_1)$ , and in 2 is  $(1 - u_2)$ ], then

$$\bar{A}_{12} = u_1 u_2 A'_{12} + u_1 (1 - u_2) A'_{10} \\ + u_2 (1 - u_1) A'_{20} + (1 - u_1) (1 - u_2) A'_{00} \dots \dots \dots (10a)$$

$$\bar{A}_{10} = u_1 A'_{10} + (1 - u_1) A'_{00} \dots \dots \dots (10b)$$

and

$$\bar{A}_{20} = u_2 A'_{20} + (1 - u_2) A'_{00} \dots \dots \dots (10c)$$

Substituting Eqs. 10a, 10b and 10c into Eq. 9 it follows that the required value is

$$\bar{A} = \frac{\bar{A}''}{n_0^2} = \frac{1}{n_0^2} u_1 u_2 (A'_{12} + A'_{00} - A'_{10} - A'_{20}) \dots \dots \dots (11)$$

From Table 1 it can be seen that  $A$  does not vary greatly with the material to be filtered, nor with the material of the filter material. Because  $A$  is nearly constant, and the filter media is usually compact material; it follows from Eq. 11 that  $\bar{A}$  is dependent almost solely on the value  $u_1$ , that is, on the density of the material to be filtered from the water, so that the constant  $A$  in the adhesion criterion can be regarded as the quantity characteristic for a given suspension.

#### CONSIDERATION OF THE CHARACTERISTIC $r$ IN THE ADHESION CRITERION

In  $\Delta r$  is the distance from each filter grain surface over which the adhesion forces are operative, there will be a volume around each grain that may be designated "adhesion space," and suspended particles entering this space will be removed from the flow as they will be attracted to adhere to the grain surface.

This they will do after traversing a distance 1. The action is analogous to a sedimentation basin of depth  $\Delta r$ , length 1, in which the gravitational force is replaced by the van der Waals force.

The proportion of the concentration removed will be equal to the proportional volume occupied by the adhesion space.

The adhesion space per unit filter volume is given by  $P \Delta r$ :

$$P \Delta r = \frac{(1 - w)^6}{D \Phi} \Delta r \dots \dots \dots (12)$$

in which  $D$  is the diameter of the filter grain,  $\Phi$  refers to the sphericity factor, allowing for the shape of the grains  $P$  denotes the specific surface of filter grains and  $w$  is the porosity.

The amount removed equals  $P \Delta r S_0$  per unit filter volume. Considering a volume element of unit plan area and depth  $(x_1 - x_0)$ ,

$$\text{Amount removed} = P \Delta r S_0 (x_1 - x_0) = \frac{(1 - w)^6}{D \Phi} \Delta r (x_1 - x_0) S_0 \dots (13)$$

But the amount removed is the difference in concentration at  $x_0(S_0)$ , and  $x_1(S_1)$ , multiplied by the volume between these levels.

$$\text{Amount removed} = w (x_1 - x_0) (S_0 - S_1) \dots \dots \dots (14)$$

Thus

$$\frac{(1-w)}{D} \frac{6}{\Phi} \Delta r S_0 = w (S_0 - S_1) \dots \dots \dots (15)$$

$$\frac{S_0 - S_1}{(x_0 - x_1)} = - \frac{(1-w)}{w} \frac{6}{D} \frac{\Delta r}{\Phi l} S_0 \dots \dots \dots (16)$$

in which

$$l = (x_1 - x_0)$$

Eq. 16 gives the change in concentration per unit depth. If the group

$$\frac{(1-w)}{w} \frac{6}{D} \frac{\Delta r}{\Phi l} = a \dots \dots \dots (17)$$

and

$$S_0 - S_1 = - a S_0 (x_0 - x_1) \dots \dots \dots (18)$$

Eq. 18 states that the change in concentration through any depth is proportional to the concentration entering that depth unit. From Eq. 17

$$\Delta r = \frac{a w D \Phi l}{(1-w)} \frac{6}{6} \dots \dots \dots (19)$$

The values on the right hand side of Eq. 19 can be measured experimentally, leading to an evaluation of  $\Delta r$ . However, the length  $l$  must be kept constant in all experiments to conserve similarity condition. This factor was chosen as a unit.

#### CONSIDERATION OF THE CHARACTERISTIC $v_r$ IN THE ADHESION CRITERION

From Eq. 8 it can be seen that

$$v_r = \frac{A}{\mu \Delta r^2 f(R)} \dots \dots \dots (20)$$

If  $\Delta r$  is determined experimentally when  $R = 1$ , that is, when  $f(R) = 1$ , then  $v_r$  can be computed. The quantity  $v_r$  is characteristic for the given suspension. Because the value  $A$  is also characteristic for the given suspension (as previously shown), it is advantageous to characterize the suspension by the fraction  $A/v_r$ , which can be experimentally determined directly from conditions when  $R = 1$ . The adhesion criterion  $Ma$  can then be determined.

$$Ma = \frac{A}{v_r \Delta r^2 \mu} \dots \dots \dots (21)$$

in which  $A$  is noted in gram centimeters squared per second,  $\mu$  is in grams per centimeter second,  $v_r$  is in centimeters per second, and  $\Delta r$  is an operational distance, in centimeters.

#### THE FILTRATION CURVE

Eq. 18 can be rewritten

$$-dS = a S dx \dots \dots \dots (22)$$

For a homogeneous filter bed and homogeneous suspension, this is an exponential relationship between concentration and depth in the filter.

$$S_x = S_0 e^{-ax} \dots \dots \dots (23)$$

For a non-homogeneous filter bed, and homogeneous suspension,

$$S_x = S_0 e^{-\int_0^x a(x) dx} \dots \dots \dots (24)$$

as  $a$  will vary with filter media size. If a curve is plotted of relative concentration  $S_x/S_0$  against depth  $x$ , as in Fig. 1, the tangent of the filtration curve at  $x = 0$  will have a slope of  $-a$ .

$$\left[ \frac{\partial \left( \frac{S_x}{S_0} \right)}{\partial x} \right]_{x=0} = - (a)_{x=0} \dots \dots \dots (25)$$

In a non-homogeneous suspension there are particles of the suspension characterized by different values of the same parameter  $s_i$ , and the value of  $a$

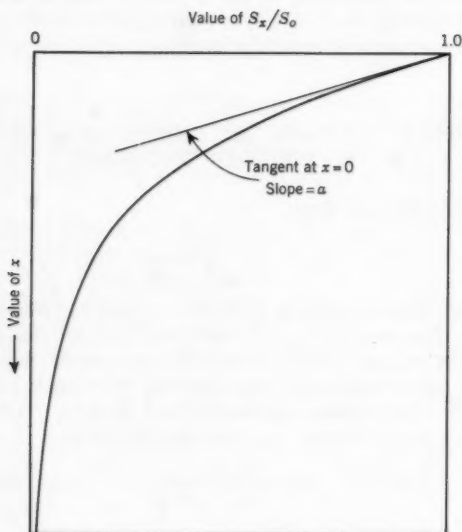


FIG. 1

depends on  $s_i$ . The suspension may be simultaneously non-homogeneous in more parameters  $s_1, s_2, \dots, s_n$  (such as density, diameter, shape, and so on). If a collection of these parameters is designated by  $s$ , the value of the concentration at any point is given then by

$$\bar{S}_x = \int_0^\infty S_x(s) ds \dots\dots\dots (26)$$

So the form of Eq. 24 becomes

$$\bar{S}_x = \int_0^\infty S_0(s) e^{-\int_0^x a(x,s) dx} ds \dots\dots\dots (27)$$

This is the generalized filtration equation of a non-homogeneous suspension in a non-homogeneous filter. Eq. 27 can be rewritten

$$\bar{S}_x = \bar{S}_0 e^{-\int_0^x a(x, \bar{s}) dx} \left\{ 1 - \frac{1}{\bar{S}_0} \int_0^\infty S_0(s) \left[ 1 - e^{-\left[ \int_0^x a(x,s) dx - \int_0^x a(x, \bar{s}) dx \right]} ds \right] \right\} \dots\dots\dots (28)$$

in which  $\bar{s}$  is the mean of the  $s$  values, so that the first part of the right hand side of the equation is true for homogeneous suspensions and the part of the equation within the brace is the correction for inhomogeneity.

The function  $a(x,s)$  can be separated into

$$a(x,s) = G(x) H(s) \dots\dots\dots (29)$$

in which  $G(x)$  is a function of  $x$  dependent on  $s$ , and  $H(s)$  is a function of  $s$  dependent on  $x$ . Eq. 28 can be transformed to

$$\bar{S}_x = \bar{S}_0 \exp \left[ - \int_0^x \bar{a} dx - \gamma_2 \left( \int_0^x \bar{a} dx \right)^2 - \gamma_3 \left( \int_0^x \bar{a} dx \right)^3 - \dots - \gamma_n \left( \int_0^x \bar{a} dx \right)^n \right] \dots\dots\dots (30)$$

in which

$$\begin{aligned} \bar{a} &= \int_0^\infty a(x,s) S_0(s) ds = G(x) \int_0^\infty H(s) S_0(s) ds = G(x) \overline{H(s)} \\ &= G(x) H(\bar{s}) \dots (31) \end{aligned}$$

and the coefficient  $\gamma$  may be expressed as an integral function of  $H(s)$  and  $S_0(s)$ .

Eq. 31 indicates the following:

1. that the tangent of the filtration curve for non-homogeneous suspension at  $x=0$  will have a slope  $-(\bar{a})_x = 0$

2. that the filtration curve may be expressed as a dependence of two dimensionless parameters  $(\bar{S}_x/\bar{S}_0)$  and  $(\int_0^x \bar{a} dx)$ . This filtration curve holds for the filtration of the given suspension through whatever filtration bed may be present.

In a homogeneous filter bed  $\int_0^x \bar{a} dx = \bar{a}x$ .

### EXPERIMENTAL TECHNIQUE

Such measurements were made on filters 100 cm deep of 10 cm-by-10 cm square plan, containing uniform sands of 1 mm and 1.2 mm diameter, and calcium carbonate grains of 1.4 mm and 1.7 mm diameter. Filtration rates of 2, 4, and 6 gpm per square ft were used for each grain size and material, and the Reynolds number for each filter was calculated from the hydraulic conditions of each. Hypodermic needles inserted 2 cm into the filter bed enabled samples to be withdrawn for analysis.

The water to be filtered and a color of 13 mg per l Platinum scale, 1 mg per l turbidity measured as silica, was dosed with either 7 mg per l  $Al_2(SO_4)_3$  or 5.5 mg per l  $FeCl_3$ , and was prepared in stirring jars. Addition of P-32 as phosphoric acid at a level of 5 microns per l was utilized to make analysis of concentration easily determined from evaporated samples counted with a G-M detector. The size of the microflocs was determined by microscopic measurement.

### EXPERIMENTAL RESULTS

By plotting the ratio of the concentration at any point to the initial concentration  $S_x/S_0$  against the dimensionless group  $ax$ , for any given suspension, the results of all the filters can be placed together on one graph. This has been done on Figs. 2 (a) and 2 (b) for  $Al(OH)_3$  floc and  $Fe(OH)_3$  floc.

These curves enable Eq. 31 to be enumerated as

$$\bar{S}_x = \bar{S}_0 \exp \left[ -\bar{a}x + 0.106 (\bar{a}x)^2 - 0.002 (\bar{a}x)^3 - 0.0002 (\bar{a}x)^4 - 0.00008 (\bar{a}x)^5 + 0.0000005 (\bar{a}x)^6 \right] \dots \dots \dots (32a)$$

for  $Al(OH)_3$  suspension, down to the limit  $\bar{S}_x/\bar{S}_0 = 0.02$ , and

$$\bar{S}_x = \bar{S}_0 \exp \left[ -\bar{a}x + 0.1196 (\bar{a}x)^2 - 0.001 (\bar{a}x)^3 - 0.0009 (\bar{a}x)^4 - 0.0000053 (\bar{a}x)^5 + 0.0000007 (\bar{a}x)^6 \right] \dots \dots \dots (32b)$$

for  $Fe(OH)_3$  suspension, down to the limit  $\bar{S}_x/\bar{S}_0 = 0.03$ .



With the value of  $\alpha$  determined empirically,  $\Delta r$  can be computed from Eq. 19 and the dimensionless group  $Ma$  calculated from Eq. 21

$$Ma = \frac{A}{v_r \Delta r^2 \mu} \dots \dots \dots (21)$$

This is shown in Table 2. Reynolds number can be calculated from the hydraulic characteristics of the filtration, and a log-log plot of  $Ma$  against  $R$  is

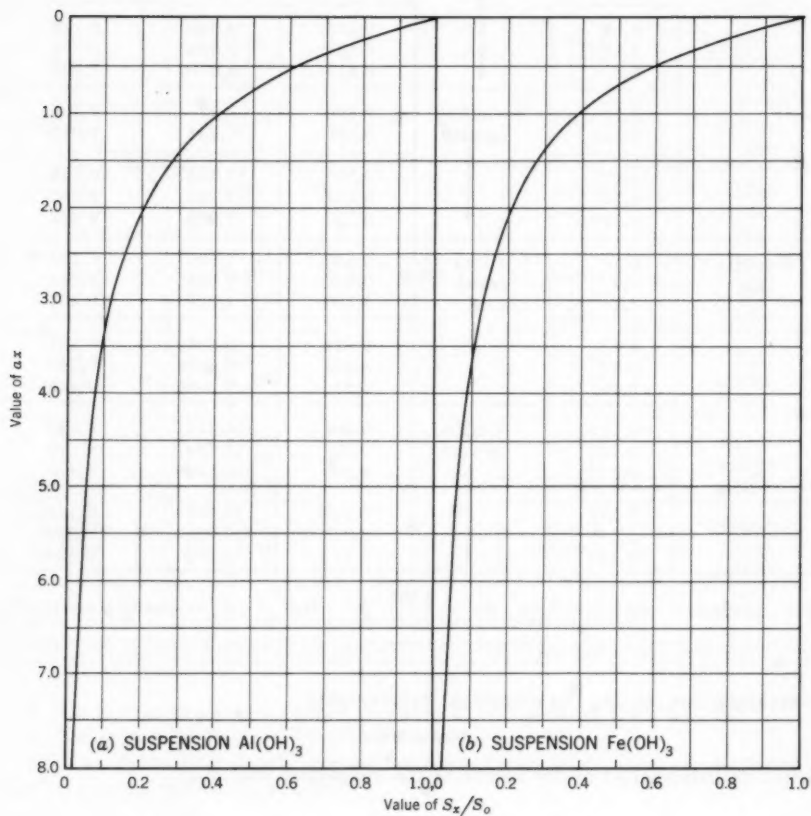


FIG. 2

TABLE 2.—COLLECTION OF

Suspension	Filter grain diameter, in centimeters	Filter medium	Porosity, w	Hydraulic radius, R in R, $\text{cm} \times 10^{-3}$	Approach velocity, V, in cm per sec
(1)	(2)	(3)	(4)	(5)	(6)
Al(OH) <sub>3</sub> floc	0.1	silica sand ↑	0.390	7.528	0.118
	0.1		0.390	7.528	0.213
	0.1		0.390	7.528	0.407
	0.12		0.428	9.0066	0.136
	0.12			9.0066	0.256
	0.12			9.0066	0.408
	0.142	CaCO <sub>3</sub> grains ↑	0.400	15.734	0.135
	0.142		0.400	15.734	0.280
	0.171		0.377	17.236	0.134
	0.171			17.236	0.272
	0.171			17.236	0.441
	Fe(OH) <sub>3</sub> floc		0.1	silica sand ↑	0.390
0.1		0.390	7.528		0.247
0.1		0.390	7.528		0.407
0.12		0.428	9.0066		0.151
0.12			9.0066		0.246
0.12			9.0066		0.438
0.142		CaCO <sub>3</sub> grains ↑	0.400	15.734	0.143
0.142			0.400	15.734	0.272
0.142			0.400	15.734	0.427
0.171			0.377	17.236	0.1424
0.171				17.236	0.2984
0.171				17.236	0.4258

For calculation:  $Ma = \frac{A}{v_r \mu \cdot \Delta r^2}$ ,  $Re = \frac{\rho VR}{\mu w}$ ,  $\Delta r = R \cdot \bar{a} \cdot 1 \text{ cm}$ , viscosity  $\mu$  and density

a straight line, giving the empirical relationship

$$Ma = R^{2.16}$$

which is independent of the nature of the suspended material.

#### APPLICATION

To determine the reduction in concentration with depth for any filter, the grading curve of which is known, the following procedure is followed:

1. Determine Reynolds number for the flow conditions
2. From Eq. 33 determine Ma

## EXPERIMENTAL RESULTS

Temperature of water, °C	Filtration parameter $\bar{a}$ , in $\text{cm}^{-1}$	Ratio $A/v_r$ , as $A/v_r \times 10^{-9}$ g, cm per sec	Adhesion criterion, Ma	Reynolds number, R
(7)	(8)	(9)	(10)	(11)
16.5	0.275	2.96	0.0631	0.2076
16.8	0.235		0.0870	0.3778
17.0	0.078		0.793	0.7259
15.5	0.243		0.0548	0.2539
15.8	0.126		0.2075	0.4822
16.0	0.077		0.556	0.7727
15.7	0.081		0.163	0.4741
17.0	0.033		1.015	1.0350
15.0	0.067		0.1903	0.5365
15.1	0.027	5.51	1.191	1.092
15.0	0.015		3.88	1.766
15.5	0.425		0.0478	0.2312
15.5	0.218		0.1823	0.4230
15.6	0.112		0.690	0.6988
18.0	0.300		0.0716	0.3011
18.0	0.193		0.173	0.4905
18.0	0.077		1.090	0.8733
17.5	0.093		0.2346	0.5126
17.5	0.052		0.751	0.9528
17.5	0.032		1.980	1.5306
17.0	0.072		0.3305	0.6015
17.0	0.032		1.740	1.2615
17.0	0.027		2.474	1.7993

of water  $\rho$  were determined from temperature using chemical tables.

- Using the value  $A/v_r$  given in Table 2, compute  $\Delta r$  from Eq. 21
- Substitute  $\Delta r$  in Eq. 19 and determine  $a$
- If  $a$  varies with depth, repeat for each depth layer (that is, each grain size, assuming hydraulic grading)
- Integrate  $\int_0^x a dx$  numerically or graphically
- Using graph 2a or 2b determine  $\bar{S}_x/\bar{S}_0$  (valid only for  $\text{Al}(\text{OH})_3$  and  $\text{Fe}(\text{OH})_3$ ; prepared under the same conditions as in the authors' experiments; other suspensions require other graphs)

From such a procedure it is possible to compare the actions of filters of different gradings, and different filtration velocities. This is illustrated in

Figs. 3 through 12. Fig. 3 shows the size distribution of sand commonly used in filters in the United States. Fig. 4 shows the coefficient  $\bar{a}$  determined for the sand grading of Fig. 6, for a filtration rate of 2 gpm per sq ft, with a porosity of 0.39, bed depth of 70 cm hydraulically stratified, operating at 15°C, with the grain sphericity factor of 0.65. Fig. 5 shows the integration of Fig. 4,

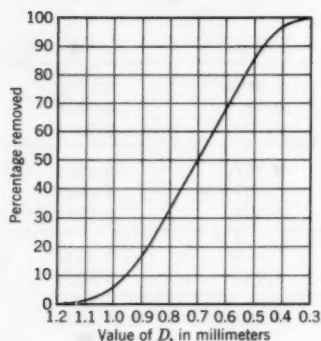


FIG. 3

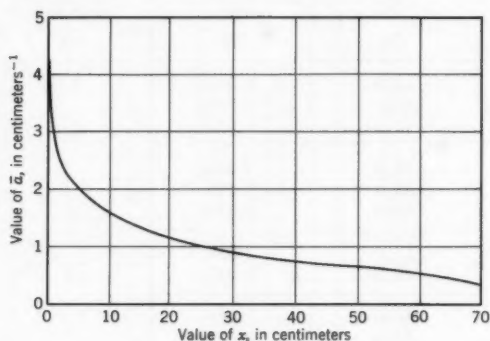


FIG. 4

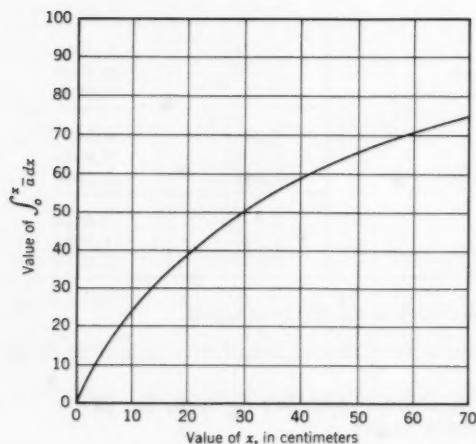


FIG. 5

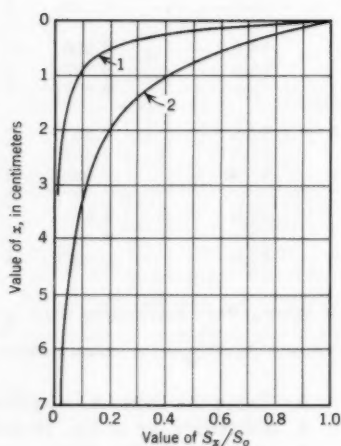


FIG. 6

$\int_0^x \bar{a} dx$ . Fig. 6 shows concentration curves, plotted from values determined from Fig. 2 (a), of  $S_x/S_0$  against depth  $x$ . Curve 1 is for the United States filter grading, curve 2 is for European filter grading, with the same total filtration efficiency. Note that the United States filter removes 90% in the top 1 cm, whereas the European filter removes 60%. European filter grain diameter

= 0.615 mm, the other parameters are the same. For both filters  $\int_0^L \bar{a} dx = 74.2$ . In Fig. 7 for a filter of uniform grain size  $D$ , the depth  $L$  is plotted that

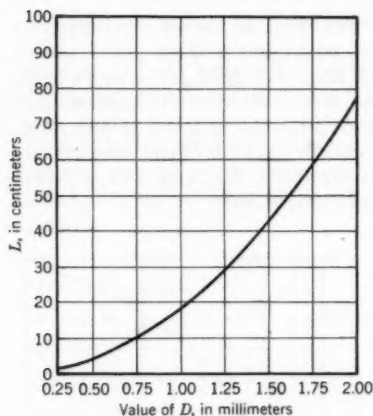


FIG. 7

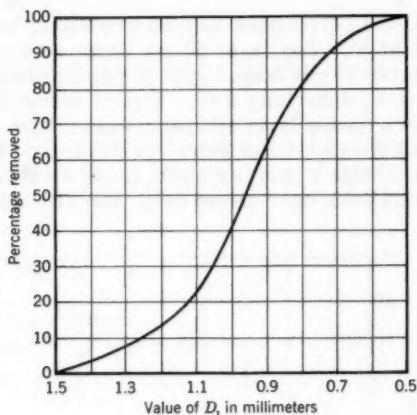


FIG. 8

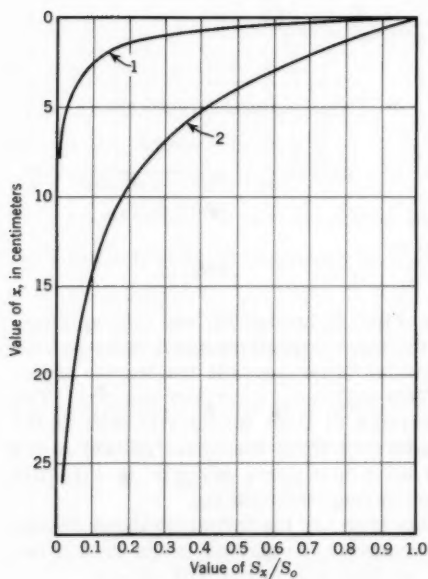


FIG. 9

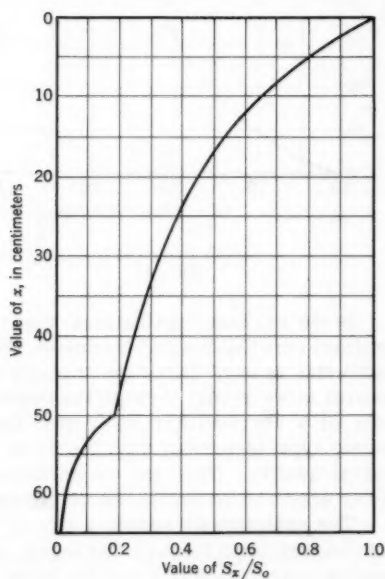


FIG. 10

will give 98% removal of suspension at a rate of 2 gpm per sq ft, 15°C, porosity 0.39, sphericity 0.65. Fig. 8 shows the size distribution of sand in a Chicago,

III. filter. Fig. 9 shows filtration through a Chicago filter, curve 1 with flow downward, curve 2 with flow upward. Rate of flow 2 gpm per sq ft, 15°C, porosity 0.39, sphericity 0.65, depth 76 cm,  $\int_0^L \bar{a} dx = 37.5$ . Note that filtration

in both directions has the same total efficiency. Fig. 10 shows two layer filtration. Top layer 50 cm deep, at 4 gpm per sq ft, grain  $D = 2$  mm. Lower layer 15 cm deep, 2 gpm per sq ft, grain  $D = 1$  mm. For both, porosity is 0.39, 15°C, sphericity 0.65. Fig. 11 shows the size distribution of the sand section of a double layer anthracite-sand filter. Anthracite uniform  $D = 0.83$  mm. Fig. 12 shows the concentration-depth curve of a double layer anthracite-sand filter, rate 4 gpm per sq ft, 15°C; anthracite 10 cm deep, porosity 0.37, sphericity 0.65; sand 38 cm deep, porosity 0.39, sphericity 0.65.

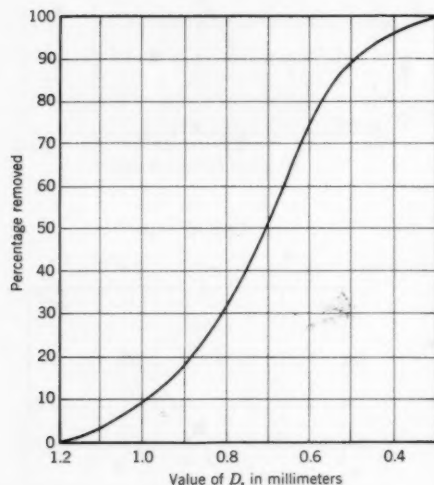


FIG. 11

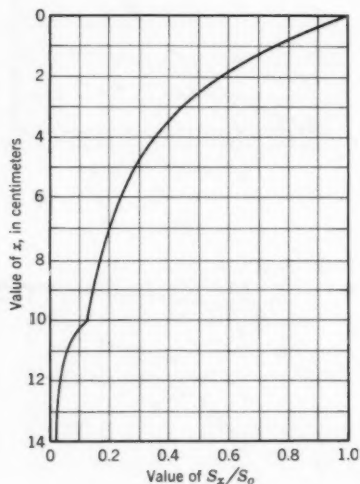


FIG. 12

In the various applications shown in Figs. 3 through 12, the following important conclusions may be drawn. For the same overall removal of suspended material upward filtration through a graded filter spreads the burden of removal more evenly through the depth. This will reduce surface clogging. The use of a coarse filter layer over the surface of finer media will lead to the same type of action, with the finer media providing the final "polish" to the water quality. This can be performed using less dense material (anthracite) over sand and so retain the configuration during backwashing.

The complete foregoing analysis holds true for the initial (in time) change of concentration through the depth. As material is retained in the sand pores so the characteristic  $\bar{a}$  and the form of the filtration curve will change.

### CONCLUSIONS

The analytical methods developed in this paper enable rational comparison of various gradings of filter media to be made and provide a basis for the fu-

ture analysis of the more complex action of the filter clogging. The fundamental approach, in linking hydrodynamic and physical chemistry phenomena, is becoming increasingly fruitful for the study of sanitary engineering processes.

#### ACKNOWLEDGMENTS

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K. J. IVES, M. ASCE, University College, University of London, London, England, who was formerly Research Fellow in Sanitary Engineering at Harvard University in Cambridge, Mass. with the permission of the authors of the original paper.

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#### APPENDIX.—NOTATION

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The following symbols, adopted for use in this paper, conform essentially with "American Standard Letter Symbols for Hydraulics" (ASA Z10.2-1942) prepared by a committee of the American Standards Association with Society representation, and approved by the Association in 1942.

A = constant of van der Waals attraction;

D = diameter of filter grain;

$D_i$  = diameter of particle;

E = Euler number;

F = Froude number;

$F_i$  = force between a particle and an infinite surface;

$F_h$  = hydrodynamic force acting on a particle in a streaming fluid;

H = Ho number;

Ma = dimensionless group  $\frac{A}{v_r r^2}$ ;

$N_o$  = refractive index of pure water;

P = specific surface of filter grains;

p = specific area of grains;

R = Reynold's number

r = distance between surface of the particle and the infinite surface;

S = concentration of suspended particles;

s = collection of parameters;

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<sup>3</sup> "Adhese ve filtracnim Lozi," by V. Mackrle and S. Mackrle, Rozpravy Ceskoslov. akademie ved, Rada tech., ved Vol. 69, No. 2, 1959.

- $v$  = velocity of fluid relative to the particle;  
 $v_r$  = particle velocity normal to surface of particle and filter grain;  
 $w$  = porosity;  
 $\gamma$  = coefficient; integral function of  $H(s)$  and  $S_0(s)$ ;  
 $\mu$  = viscosity of fluid;  
 $\rho$  = density of fluid; and  
 $\Phi$  = sphericity factor, allowing for the shape of the grain.



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REFUSE-SLUDGE COMPOSTING IN WINDROWS AND BINS

By John S. Wiley,<sup>1</sup> F. ASCE, and Janet T. Spillane<sup>2</sup>

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SYNOPSIS

Successful composting was achieved at Chandler, Ariz., in two runs involving 4 windrows and 25 aeration bins of ground refuse and refuse-sludge mixtures. The composting materials were turned an average of 5.1 times by re-grinding and were composted for a mean of 34.4 days. The two methods produced very similar results with an average decomposition of 38.8% of the volatile solids. The raw refuse was of poor quality and the sludge had a high water content. Pasteurizing temperatures were maintained for days, and there were little odor or fly problems. The windrow method appears to be practical for plant-scale use and should adequately decompose refuse or refuse-sludge mixtures in a total time of about 5 weeks with five vigorous turnings during the first 3 weeks.

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Note.—Discussion open until February 1, 1962. To extend the closing date one month, a written request must be filed with the Executive Secretary, ASCE. This paper is part of the copyrighted Journal of the Sanitary Engineering Division, Proceedings of the American Society of Civil Engineers, Vol. 87, No. SA 5, September, 1961.

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## INTRODUCTION

The grinding plant at Chandler, Ariz., as described by Maier, Williams, and Mallison<sup>3,4</sup> was used for preparation and handling of refuse and refuse-sludge mixtures for experimental outdoor composting by the windrow method and the aeration-bin method.<sup>4</sup> Refuse from the City of Chandler was grossly sorted with a few large objects and rags being picked out and with ferrous metals removed by an electromagnet that operated at about 60% efficiency.<sup>4</sup> The refuse was ground through a hammermill with coarse bar grate, water or sludge being added at the bottom of the primary elevating conveyor.

Two runs were made during the period from July 8 to August 23, 1958. Run 1 comprised three groups each with a 4 to 6-ton windrow and seven aeration bins also containing 4 to 6 tons. Groups 1 and 2, with Windrows 1 and 2 and Bins 1-14, contained refuse and water. Group 3, with Windrow 3 and Bins 15-21, contained approximately 71% refuse and 29% raw sewage sludge (by weight). Run 2 comprised one 2.7-ton windrow and four bins containing 2.8 tons. These units, Windrow 1 and Bins 1-4, contained 18.4% and 28.2% sludge (wet weight), respectively. The raw sludge was pumped to a holding tank from the nearby Chandler sewage treatment plant.

The outdoor composting was conducted in windrows about 4 ft high and 6 ft wide at the base, on concrete slabs; and in hardware cloth bins 30 in. by 72 in. by 48 in. high (60 cu ft) exposed to the air on all six sides. The active composting was conducted for 14 or more days with turning at intervals. Turning consisted, in every case, of running the material through the entire plant, wetting, and replacing it. Water also was occasionally sprinkled over the surfaces of the material in the windrows and bins. In some cases, a group was piled or windrowed for curing without further turning or handling. Because the turnings involved regrinding, each turning is called a "regrind" herein. There were from three to eight regrinds for each group. Average time from initial grinding to harvesting was 34.4 days.

The field observations and analyses were made at the plant where a trailer laboratory was located. These consisted of temperature, total solids, moisture, pH, and oxygen determinations; observations on color, odor, and fly breeding in the composting matter; and measurements of total weight and volume. Laboratory analyses, conducted in Savannah, Ga., consisted of determination of volatile solids, ash, lipids (ether soluble matter), crude fiber, nitrogen, protein (calculated from N content), carbon, and C/N value.<sup>5</sup>

Table 1 shows the composting schedule for all groups in both runs, giving with respect to time the regrinds, weighings, moisture samples, samples for more complete analyses, stacking for curing, and harvesting of the compost.

<sup>3</sup> "Composting Studies. I. Composting Municipal Refuse by the Aeration Bin Process," by R. P. Maier, E. R. Williams, and G. F. Mallison, Proceedings, 12th Industrial Waste Conf., Purdue Univ., Lafayette, Ind., Engrg. Bulletin, Vol. 42, No. 3, September, 1958, p. 590.

<sup>4</sup> "Grinding Stations to Minimize Refuse Handling Costs," by R. P. Maier, E. R. Williams, and G. F. Mallison, manuscript.

<sup>5</sup> "Methods for Examination of Raw and Composted Organic Wastes," Tech. Development Labs., CDC, PHS, Chem. Memo. No. 4, mimeo., December 3, 1956.

TABLE 1.—COMPOSTING SCHEDULE

Days	Run 1						Run 2	
	Group 1		Group 2		Group 3		Group 1	
	Windrow 1	Bins 1-7	Windrow 2	Bins 8-14	Windrow 3	Bins 15-21	Windrow 1	Bins 1-4
0	WS	WS	WS	WS	WS	WS	WS	WS
1								
2							R-1	R-1
3								
4	R-1 W							
5		M	R-1 W		R-1 WM	R-1 WM		
6		R-1 WS		R-1 W S			R-2	R-2
7	S		S	S	R-2 WS	R-2 WS		
8			R-2 W		M	M		R-3
9	R-2 W	R-2 WM		R-2 WM			R-3 S	S
10				M	R-3 M	R-3 M	R-4	
11								R-4 M
12	M	M	R-3 M				M	
13								
14	R-3 WCS	R-3 WCS	R-4 WS	R-3 WS	S	R-4 WCS		
15								R-5 WS
16							R-5 WS	
17					R-4 WCM			
18								
19			R-5 S	R-4				
20								
21							R-6 WCS	R-6 WCS
22			R-6	R-5				
23								
24								
25								
26								
27								
28		M						
29				R-6 WS	HWS	HWS		
30		HWS	R-7 WS	M				
31	HWS							
32								
33								
34								
35							HWS	HWS
36				R-7 M				
37			R-8 WM					
38								
39								
40								
41								
42								
43			R-9 HWS	R-8 HWS				

Legend: W - Weighed; S - Sample for complete analysis; M - Sample for moisture determination; R - Re grind;  
2 - Number of Re grind; C - Piled for curing; H - Harvested.

In some cases, small piles of compost were held for further observations of temperature, moisture, and appearance beyond the time of harvesting the bulk of the material.

### RAW MATERIALS

The mixed raw refuse from primarily residential and light commercial sections was delivered to the plant in the regular packer trucks operated by the City of Chandler. Lack of manpower prevented adequate sorting for maximum removal of salvage and of non-compostable matter. Ferrous metals removed amounted to about 5.6% of the total wet weight; rags, discards, and moisture losses during storage, sorting, and grinding amounted to about 11%. Steps in initial preparation included dumping the refuse on a concrete apron, pushing it into the primary conveyor with a tractor, gross hand sorting, liquid addition, grinding, secondary conveying, electromagnetic separation, and placing of the ground raw products in bins or windrows.

To secure representative samples of the raw refuse, it was necessary to subject it to grinding and mixing. Therefore, the regular sorting, grinding, and iron removal were done on a small portion of the raw refuse but without the addition of water or sludge. Small samples then were obtained after repeated mixing and quartering. As a final check, the weights of ash were calculated for each sub-group several times during the run, when both total weights and analytical analyses were made. The weight of ash for a run should not change appreciably. Therefore, when there was appreciable change, it was assumed that the raw samples were not truly representative and the analyses were adjusted by correcting the ash content so that the total ash weights showed no significant change. Samples of compost showed less variation due to the numerous regrinds, which reduced particle size and accomplished good mixing. Schulze made use of constant ash weights during composting to calculate weights of other constituents from daily analyses during composting.<sup>6</sup>

In some of the tables that follow, constituent percentages are shown on a volatile solids (ash-free) basis. Most of the ash content in United States refuse arises from glass, ceramics, metals, and similar substances that are nondecomposable. When refuse is only very grossly sorted, as in these runs, a high proportion of inorganic materials exists in the materials to be composted. Computation of analyses on an ash-free basis expresses the constituents roughly in terms of the dry organic matter, rather than on a total solids basis, and gives another basis for comparison. For example, the refuse at Savannah appeared to be quite similar to that at Chandler. However, in composting experiments in laboratory units at Savannah, very complete sorting was utilized and practically all glass, ceramics, metals, and similar materials, as well as about half the paper, were removed prior to grinding and composting.<sup>7</sup> The result was an average ash content of only 7.8% of the dry weight, compared to the 33.0% average for the grossly sorted Chandler refuse. Table 2 compares the analyses of Savannah and Chandler refuse as sorted and ground ready for composting.

<sup>6</sup> "Aerobic Decomposition of Organic Wastes," by K. L. Schulze, Michigan State Univ., East Lansing, Mich., Interim Report No. 3, January 15, 1958.

<sup>7</sup> "A Preliminary Study of High-Rate Composting," by J. S. Wiley and G. W. Pearce, *Transactions, ASCE*, Vol. 122, 1957, p. 1009.

TABLE 2.—COMPARISON OF ANALYSES OF RAW REFUSE PREPARED FOR COMPOSTING AT SAVANNAH, GEORGIA, AND CHANDLER, ARIZ.

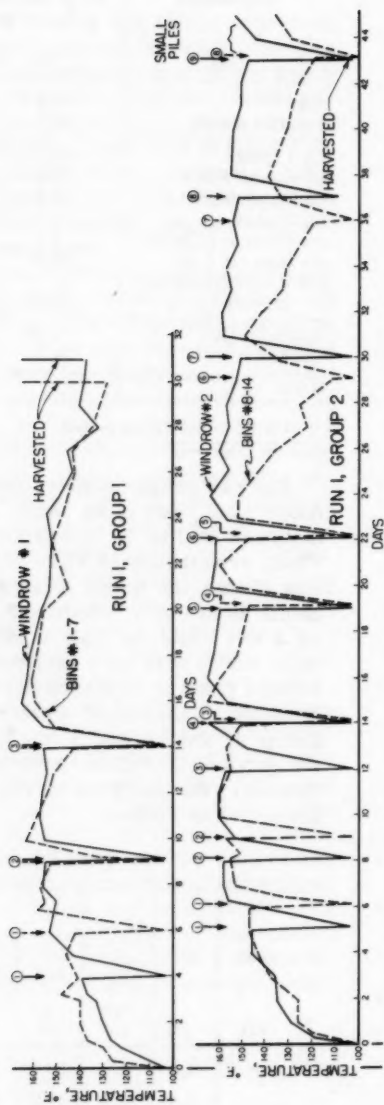
Constituent  (1)	Percentage of Dry Weight			
	Savannah Refuse (well-sorted)		Chandler Refuse (grossly sorted)	
	Total Solids Basis (2)	Volatile Solids (Ash-free) Basis (3)	Total Solids Basis (4)	Volatile Solids (Ash-free) Basis (5)
Ash	7.9	....	33.0	....
Volatile Solids	92.1	100.0	67.0	100.0
(1) Lipids	10.2	11.1	6.6	9.8
(2) Crude Fiber	35.6	38.7	37.3	55.7
(3) Total Sugar	5.0	5.5	N.D. <sup>a</sup>	N.D. <sup>a</sup>
(4) Starch	8.3	9.0	N.D. <sup>a</sup>	N.D. <sup>a</sup>
(5) Protein (6.25 N)	8.1	8.8	3.6	5.3
(6) Sum (1) - (5)	67.2	73.1	47.5	70.8
(7) V.S.-(6)(Undetermined)	24.9	26.9	19.5	29.2
Nitrogen (N)	1.30	1.41	0.57	0.85
Carbon (C)	50.1	54.4	37.5	56.0
Ratio: C/N	38.5		65.8	

<sup>a</sup> N.D. - Not Determined.

The raw sewage sludge, obtained from the primary clarifiers of a trickling filter plant, was quite weak. It contained 98.6% moisture in Run 1 and 98.8% moisture in Run 2, compared to the usual estimate of 95% to 96% moisture. Thus, even though 18.4% to 30.2% of the wet weight of refuse-sludge mixtures was sludge, the weight of sludge on a dry basis was almost negligible. An example is in Run 1, Windrow 3, which contained 29.1% sludge and 70.9% refuse on a wet basis but only 0.84% sludge and 99.16% refuse on a dry basis. The weak sludge may have been due to faulty sludge collection or withdrawal at the sewage plant or to inadequate mixing in the holding tank at the compost plant. Because of the small amount of sludge solids added to the refuse in Run 1, group 3, and in Run 2, the composting was essentially that of refuse alone. All groups, therefore, have been combined in some of the tables that follow. However, the analyses of the raw sludge solids might be of interest and are presented in Table 3.

TABLE 3.—ANALYSES OF RAW SLUDGE SOLIDS

Run  (1)	pH  (2)	Mois- ture, per- cent- age (3)	Percentage of Dry Weight							Ratio C/N  (11)
			Ash  (4)	Vol. Sol.  (5)	Crude Fiber  (6)	Lip- ids  (7)	Pro- tein (6.25N)  (8)	Nitro- gen  (9)	Car- bon  (10)	
1	6.4	98.6	26.8	73.2	22.1	14.2	12.2	1.95	35.4	18.2
2	5.8	98.8	31.7	68.3	12.0	12.6	15.5	2.48	43.3	17.5



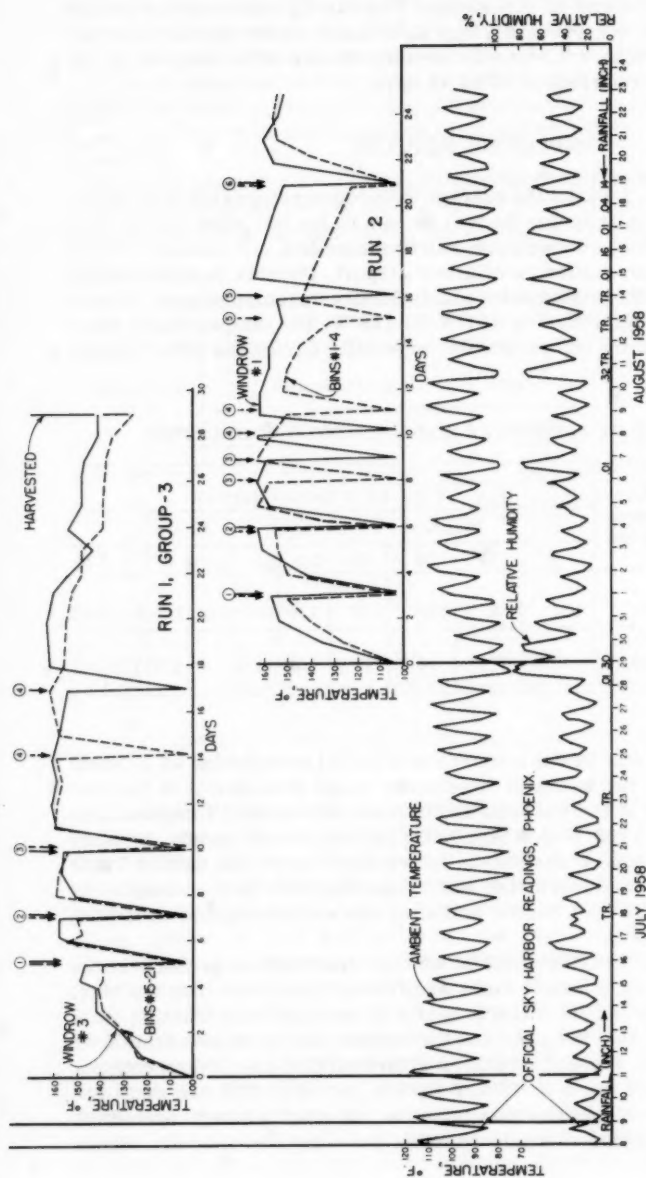


FIG. 1.—TEMPERATURE AT 10-IN. DEPTH IN REFUSE AND REFUSE-SLUDGE COMPOST



The addition of raw sludge to refuse for composting should speed up the decomposition and improve the value of the end product, as the sludge has a lower C/N and a higher N content than refuse. A considerably higher N content was expected than was found in the sludge. The raw Chandler refuse had an extremely low nitrogen content and a high C/N value, which may account for continuing decomposition as evidenced by the temperature of the compost at the end of the decomposition periods of 29 to 43 days.

### COMPOSTING RESULTS

*Temperatures.*—Fig. 1 shows the average daily temperatures taken at 10-in. depth in the composting matter for the four groups in the two runs. At the bottom are shown the ambient temperature, relative humidity, and rainfall occurring at the official weather station, Sky Harbor Airport, Phoenix (approximately 12 miles from the Chandler composting plant). Maximum temperatures in each subgroup were within the range of 161°F to 165°F. Bin temperatures were slightly lower than windrow temperatures, especially during the latter stages

TABLE 4.—RAPID TEMPERATURE RISE AND OXYGEN DEPLETION

Measurement	Before Second Regrind	Time After Second Regrind									
		Minutes				Hours					
		5	30	50	80	2.5	3	3.5	4	5	20
Temperature, in °F at 10-in. depth	155	104	108	110	110	110	113	122	118	132	163
Oxygen, in % by vol- ume at 15-in. depth	10.1	18.6	17.7	14.7	11.4	9.0	11.4	1.6	2.7	1.2	6.4

of decomposition. This may be due to more rapid initial composting as a result of better aeration, or it may be due to the smaller mass of material in the bins and greater desiccation of the material as the mass decreased. Temperatures dropped to about 100°F to 103°F upon regrinding but recovered rapidly, in many cases to a higher value than immediately before regrinding. The data in Table 4 illustrate the rapid temperature rise and oxygen depletion in the compost as measured in Run 2, Bins 1-4 before and after the second regrind on the 6th and 7th days of composting.

Temperature varied with the depth at which it was taken especially in the windrows which had a larger single mass and greater insulation than the bins. However, it was not only the insulating property of the mass that changed temperature with depth but also the time and the oxygen content after aeration or regrinding. For example, the temperatures at three depths and oxygen concentrations at two depths are given in Table 5 for the period of fifth and sixth regrinds of Windrow 1, Run 2. These data indicate that shortly after regrinding, the temperatures are higher at shallow depths; subsequently, they are higher at greater depths.



At the end of curing the compost from Windrow 1, Run 1 (composting with three regrinds in 14 days, followed by curing for 17 days undisturbed), the windrow was sectioned. There were four different-appearing zones with quite different characteristics. These had temperatures as follows: top zone, 3-8 in. thick, 132°F; second zone, 12-in. to 15-in. thick, 160°F; third zone, 30-in. to 32-in. thick, 160°F; and bottom zone, 0-in. to 3-in. thick, 137°F. Lower temperatures appearing in outer zones on standing may be due to reduced decomposition because of partial drying and poor insulation. Higher central temperatures may be due to a combination of continued active decomposition and good insulation.

*Oxygen Concentrations.*—Oxygen concentrations were measured by inserting a small-bore probe to the desired depth in the bin or windrow and drawing a sample therefrom through an oxygen analyzer. The concentration was read in

TABLE 5.—TEMPERATURES AT THREE DEPTHS AND OXYGEN CONCENTRATIONS AT TWO DEPTHS

Composting day	Temperature, in °F at depth:			Oxygen, in % by volume at depth:	
	6-in.	10-in.	24-in.	12-in.	24-in.
16	146	154	...	6.0	1.9
16	Fifth Regrind				
17	161	164	141	1.2	1.0
18	157	162	154	3.2	1.0
20	147	155	149	6.6	1.2
21	139	151	151	7.0	1.0
21	Sixth Regrind				
22	153	156	117	1.2	1.2
23	154	160	148	2.6	0.8
24	147	154	142	7.0	1.2
25	140	151	151	7.4	1.3

mm partial pressure of O<sub>2</sub> and converted to percentage by volume from a calibration curve. Normal air at 760 mm pressure and 70°F has an oxygen content of about 21% and an O<sub>2</sub> partial pressure of about 159 mm. Shortly after grinding or regrinding, the atmosphere within a bin or windrow was found to contain 18.6% to 19.0% oxygen. Normally, readings were made at 12-in. and 15-in. depths in bins and at 12-in. and 24-in. depths in windrows. By depth here is meant distance from the nearest surface exposed to the air, whether the distance is horizontal, vertical, or at an angle. In a bin, the greatest distance possible was 15 in. as the bins were 30 in. wide.

Fig. 2 shows oxygen contents at 15-in. depth in bins and 24-in. depth in windrows, and at 12-in. and 24-in. depths in curing piles for Run 1, groups 1 and 3. By the 4th to 6th day, the oxygen level dropped to 2% or less in the windrows and remained at that level until the 21st day or later. In group 2 (not shown), the oxygen level in windrow 2 at 24-in. depth was still below 2% after 45 days of composting and nine regrinds. The bins usually showed extremely

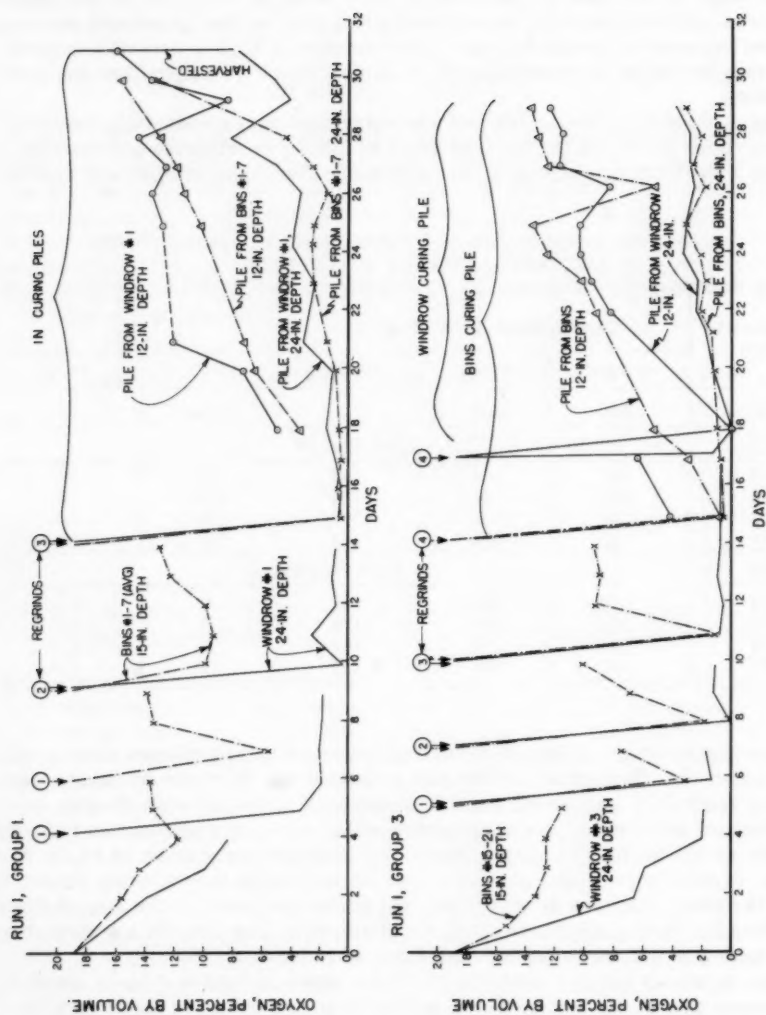


FIG. 2.—OXYGEN IN REFUSE COMPOST

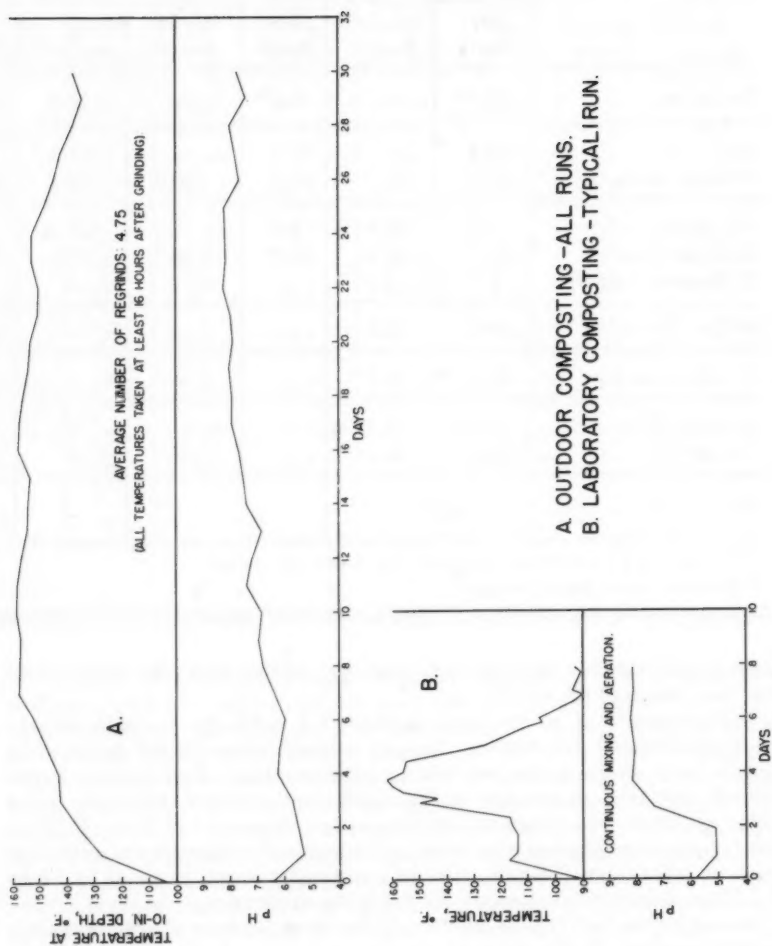


FIG. 3.—TEMPERATURE AND pH OF COMPOST

TABLE 6.—CHANGES IN ANALYSES AND WEIGHTS WITH TIME OF COMPOSTING,

Constituents	0-day		7-day			Analys Dry Basis
	Analyses, percent		Analyses, percent		Change in Wt., percent	
	Dry Basis	Vol. Sol. Basis*	Dry Basis	Vol. Sol. Basis*		
Dry Solids	55.8 x	—	49.8 x	—	-12.4	48.9 x
Ash	33.3	—	39.3	—	+3.5	39.6
Volatile Solids	66.7	100.0	60.7	100.0	-20.2	60.4
(1) Lipids	7.3	10.9	5.0	8.3	-39.3	2.4
(2) Crude Fiber	35.3	52.8	35.9	59.1	-10.8	34.6
(3) Protein (6.25 N)	3.7	5.6	—	—	—	3.5
(4) Sum (1) – (3)	46.3	69.3	—	—	—	40.5
(5) V.S. – (4) (Undet.)	20.4	30.7	—	—	—	19.9
Nitrogen (N)	0.59	0.89	—	—	—	0.57
Carbon (C)	37.0	55.5	—	—	—	35.2
Ratio: C/N	62.4		—		—	

\* Percent on a volatile solids (ash-free) basis, by weight.

<sup>x</sup> Percent of wet (total) weight.

low oxygen contents only the first day after regrinding; then, the oxygen content rose to a range of 6% to 14%.

The initial period up to the first grind (3rd to 6th day in these experiments) is particularly interesting. Oxygen concentration slowly declined in contrast to later sharp decreases following the regrinds. Also, temperatures rose slowly initially, in contrast to the rapid rises following regrinds; there was some evidence of a temperature plateau in a range of 126°F to 145°F. A similar temperature plateau was found in mechanical composting of refuse at Savannah in laboratory (15-gal) units in a somewhat lower range of 110°F to 120°F.<sup>7</sup> This plateau was believed to be due to the predominant action of mesophilic, acid-forming bacteria in advance of the establishment of predominantly thermophilic action. In the case of the larger masses in bins and windrows at Chandler, the higher temperature plateau may be due to greater insulation and higher ambient temperatures (average ambient temperature at Chandler was 94°F). The initial three days of decomposition generally did not reduce the oxygen level below about 6%.

*pH Values.*—The pH values were measured by potentiometer daily from small grab samples taken at 8-in. to 12-in. depth. These varied widely; it was

RUN 1, WINDROW 2 AND BINS 8-14 (AVERAGE VALUES FOR RUN 1)

14-day		30-day			43-day		
es, percent	Change in Wt., percent	Analyses, percent		Change in Wt., percent	Analyses, percent		Change in Wt., percent
		Dry Basis	Vol. Sol. Basis*		Dry Basis	Vol. Sol. Basis*	
—	-18.1	58.1 <sup>x</sup>	—	-29.9	68.3 <sup>x</sup>	—	-35.1
—	- 2.6	47.0	—	- 5.5	48.5	—	- 5.5
100.0	-25.9	53.0	100.0	-44.3	51.5	100.0	-49.9
4.0	-72.6	1.8	3.3	-83.0	1.5	2.9	-86.7
57.3	-19.6	28.3	53.5	-43.7	27.8	53.9	-48.9
5.9	-22.1	3.9	7.3	-26.4	4.5	8.8	-20.8
67.2	-28.1	34.0	64.1	-48.5	33.8	65.6	-52.6
32.8	-20.8	19.0	35.9	-34.9	17.7	34.4	-43.7
0.94	-22.1	0.62	1.18	-26.4	0.72	1.40	-20.8
58.3	-22.1	30.0	56.7	-43.1	28.4	55.1	-50.3
62.4	—	48.2		—	39.2		—

not learned until late in the experiments that pH varied greatly with the depth. However, average values for all runs and groups are shown in Fig. 3, along with compost temperatures. For comparison a typical run in a laboratory mechanical unit also is shown. Except in Run 2, Windrow 1, the pH did not reach and remain above 7.0 until the 8th day of composting. However, temperatures above 150° were attained and maintained when the pH rose to a value of about 6.4. There apparently was a considerable period, from about the 3rd to the 7th day, when both mesophiles and thermophiles were quite active with the latter gradually becoming predominant.

As an example of the difference in pH with depth, reference again is made to the sectioning of the curing pile in Run 1, Windrow 1, after 31 days of decomposition. In the four different zones from top to bottom, the pH values were as follows: 7.9, 5.9, 5.5, and 7.5. The pH of the mixture was 6.6. The two high pH values were found in gray zones in which it appeared that actinomycetes were the predominant organisms. These were the outer and bottom zones, which were also the driest and the coolest.

On the basis of the observation that as composting becomes more and more complete the pH gradually approaches 7.0 (from higher values), Fig. 3 shows

that decomposition was not completed but was approaching completion. The relatively high temperatures at the conclusion of the runs, however, indicate that appreciable active decomposition may still have been occurring.

*Chemical Analyses.*—Chemical analyses with respect to time are shown in Table 6 for the longest composting run (Run 1, group 2 involving Windrow 2 and Bins 8-14). Over-all average values are reported based on averages from Windrow 2 and averages from Bins 8-14. Decomposition proceeded most rapidly in the case of lipids (oils, fats, and waxes). The rate of lipid decomposition declined after the 14th day. Volatile solids, carbon, crude fiber, and the undetermined volatiles (Item 5) showed steady decomposition through the 30th day, then at a slightly lesser rate. More nitrogen (and also protein) was lost than anticipated. However, the summary (Table 9) indicates less N loss than

TABLE 7.—COMPARISON OF WINDROW AND BIN COMPOSTING WEIGHTS AND WEIGHT CHANGES IN POUNDS.

Constituents (1)	All Windrows			All Bins		
	Raw (2)	Compost (3)	% Change (4)	Raw (5)	Compost (6)	% Change (7)
Total Wet Weight	36,185	21,218	-41.4	34,895	21,264	-39.1
Moisture	16,772	7,187	-57.1	16,375	7,149	-56.3
Dry Solids	19,413	14,031	-27.7	18,520	14,115	-23.8
Ash	6,321	6,180	- 2.2	6,190	6,406	+ 3.5
Volatile Solids	13,092	7,851	-40.0	12,330	7,709	-37.5
(1) Lipids	1,297	339	-73.9	1,203	251	-79.1
(2) Crude Fiber	7,303	4,464	-38.9	6,845	4,424	-35.4
(3) Protein (6.25N)	699	550	-21.4	654	581	-11.1
(4) Sum (1) - (3)	9,299	5,353	-42.4	8,702	5,256	-39.6
(5) V.S.-(4) (Undetermined)	3,793	2,498	-34.1	3,628	2,453	-32.4
Nitrogen (N)	111.9	88.0	-21.4	104.6	93.0	-11.1
Carbon (C)	7,314	4,672	-36.1	6,921	4,352	-37.1
Ratio: C/N	65.4	53.1	-18.8	66.2	46.8	-29.3

shown in Table 6. The C/N value was high initially and was not reduced sufficiently to indicate stability. For horticultural or agricultural use, the final compost was considerably higher in ash and C/N value and lower in N content than desired. Considerable improvement could have been provided by better sorting of raw refuse to remove more glass, metals, ceramics, and paper. This sorting would have reduced initial ash and C/N values and increased the initial per cent nitrogen.

Comparison of bin and windrow composting for all runs is shown by Tables 7 and 8. The weights in Table 7 show that approximately the same total quantities were treated by the two methods, although there were only 4 windrows compared to 25 bins. Regarding treatment during composting, the windrow materials were composted and cured for an average of  $34\frac{1}{2}$  days with an

average of  $5\frac{1}{4}$  regrinds, while the bin materials were composted and cured  $34\frac{1}{4}$  days with 5 regrinds. Each group was treated as nearly the same as possible, except that the bins were sprinkled more frequently than the windrows due to greater moisture evaporation.

From the losses in total and volatile solids, crude fiber, and undetermined organic matter (item 5), it appears that the decomposition in windrows was greater than that in bins, but probably not by significant amounts. However, the bin contents lost less nitrogen and showed greater reductions in lipids and C/N values than did the windrow contents. The weights indicate good de-

TABLE 8.—COMPARISON OF WINDROW AND BIN COMPOSTING ANALYSES

Constitu- ents  (1)	Percentage by Dry Weight							
	All Windrows				All Bins			
	Raw		Compost		Raw		Compost	
	Dry Basis (2)	Vol. Sol. Basis <sup>a</sup> (3)	Dry Basis (4)	Vol. Sol. Basis <sup>a</sup> (5)	Dry Basis (6)	Vol. Sol. Basis <sup>a</sup> (7)	Dry Basis (8)	Vol. Sol. Basis <sup>a</sup> (9)
Dry Solids	53.6 <sup>b</sup>	...	66.1 <sup>b</sup>	...	53.1 <sup>b</sup>	...	66.4 <sup>b</sup>	...
Ash	32.6	...	44.0	...	33.4	...	45.4	...
Volatile Solids	67.4	100.0	56.0	100.0	66.6	100.0	54.6	100.0
(1) Lipids	6.7	9.9	2.4	4.3	6.5	9.8	1.8	3.3
(2) Crude Fiber	37.6	55.8	31.8	56.9	37.0	55.5	31.3	57.4
(3) Protein (6.25N)	3.6	5.3	3.9	7.0	3.5	5.3	4.1	7.5
(4) Sum (1) - (3)	47.9	71.0	38.1	68.2	47.0	70.6	37.2	68.2
(5) V.S.-(4) (Undetermined)	19.5	29.0	17.9	31.8	19.6	29.4	17.4	31.8
Nitrogen (N)	0.58	0.85	0.63	1.12	0.56	0.85	0.66	1.21
Carbon (C)	37.7	55.9	33.3	59.5	37.4	56.1	30.8	56.5
Ratio: C/N	65.4		53.1		66.2		46.8	

<sup>a</sup> Percentage on a volatile solids (ash-free) basis, by weight

<sup>b</sup> Percentage on a wet (total) weight basis

composition by both methods and there appears to be little choice between the two from this standpoint.

The comparative analyses in Table 8 indicate the same trends as shown by the gross weights. Here again, initial ash contents and C/N values are higher and nitrogen content is lower than desired. Paper was apparently decomposed (as indicated by crude fiber content) as readily as all other carbonaceous matter.

Tables 9 and 10 present average values for all runs at Chandler and for a number of runs in laboratory units at Savannah for comparison. In addition to analyses and weight losses, the tables show whether or not there were



significant differences between the initial (raw) and final (compost) values at the 95% level by means of the t-test.<sup>8</sup> In the case of Table 9, the weights are grand totals, whereas in Table 10 the weights are average values by laboratory composter.

Smaller nitrogen losses were encountered in the more rapid laboratory composting than in the longer outdoor composting; the latter was significant at about the 96% level. While the raw refuse at Savannah was better sorted and of better quality (less ash, lower C/N, higher N%) than the Chandler refuse, the analyses are comparable if considered on an ash-free basis. In this case, the main difference is the higher paper content of Chandler refuse due to less complete sorting of paper and cardboard. Sugar and starch determinations

TABLE 9.—SUMMARY OF ANALYSES AND WEIGHT CHANGES IN OUTDOOR COMPOSTING OF REFUSE.<sup>a</sup>

CONSTITUENT	RAW MIXTURE* PERCENT BY WT.		COMPOST PERCENT BY WT.		SIGNIF. DIFF. AT 95% LEVEL <sup>x</sup>	WEIGHT, LB.		CHANGE IN WEIGHT %	SIGNIF. DIFF. AT 95% LEVEL
	DRY BASIS	VOL. SOL. BASIS	DRY BASIS	VOL. SOL. BASIS		INITIAL	FINAL		
TOTAL WET WT.	100.0 †	—	100.0 †	—	—	71,080*	42,482	-40.2	YES (-)
MOISTURE	46.6 †	—	33.7 †	—	YES (-)	33,147*	14,336	-56.8	YES (-)
TOTAL SOLIDS	100.0	—	100.0	—	—	37,933	28,146	-25.8	YES (-)
ASH	33.0	—	44.7	—	YES (+)	12,511	12,586	+ 0.6	NO
VOLATILE SOLIDS	67.0	100.0	55.3	100.0	YES (-)	25,422	15,560	-38.8	YES (-)
(1) LIPIDS	6.6	9.8	2.1	3.8	YES (-)	2,500	590	-76.4	YES (-)
(2) CRUDE FIBER	37.3	55.7	31.6	57.1	YES (-)	14,148	8,888	-37.2	YES (-)
(3) PROTEIN (6.25N)	3.6	5.3	4.0	7.3	YES (+)	1,353	1,131	-16.4	YES (-)
(4) SUM (1) TO (3)	47.5	70.8	37.7	68.2	YES (-)	18,001	10,609	-41.1	YES (-)
(5) VS-(4) UNDET.	19.5	29.2	17.6	31.8	YES (-)	7,421	4,951	-33.1	YES (-)
NITROGEN (N)	0.57	0.85	0.64	1.16	YES (+)	2165	181.0	-16.4	YES (-)
CARBON (C)	37.5	56.0	32.1	58.0	YES (-)	14,235	9,024	-36.6	YES (-)
RATIO: C/N	65.8		49.9		YES (-)	65.8	49.9	-24.2	YES (-)

\*PARTIALLY SORTED AND GROUND REFUSE (WATER ADDED) IN 2 WINDROWS AND 14 BINS; REFUSE WITH AN AVERAGE OF 27.3 % RAW SEWAGE SLUDGE (WET WT.) IN 2 WINDROWS AND 11 BINS.

<sup>x</sup> CALCULATED FROM PERCENT DRY WEIGHT VALUES.

† PERCENT OF WET (TOTAL) WEIGHT.

<sup>a</sup> DOES NOT INCLUDE MOISTURE ADDED DURING COMPOSTING RUNS.

<sup>a</sup> AVERAGE VALUES BASED ON 4 WINDROWS AND 25 BINS (CHANDLER) (MEAN NUMBER OF REGRINDS, 5.1, MEAN COMPOST TIME, 34.4 DAYS).

were not made in the case of the Chandler experiments, but one would surmise that virtually complete removal of these constituents would have been attained in the outdoor composting as they were in laboratory composting<sup>7</sup> (Table 10). There is some evidence (Table 9) to support this assumption, since the undetermined volatile matter (item 5) showed a greater reduction than that in Table 10 (item 7). Moisture losses, which are also reflected in total wet weight losses, are of no importance in evaluating degree of decomposition since they

<sup>8</sup> "Statistical Methods," by G. W. Snedecor, Iowa State College Press, Ames, Iowa, 5th Edition, 1956.



indicate only the amount of evaporation that occurred. Evaporation was much greater than indicated by the raw and compost weights due to rains and intermittent wetting of the materials. Air drying or further curing of the compost under shelter would reduce moisture considerably more in most United States climates and would result in overall weight reductions of half or more.

*Weight and Volume Measurements.*—Weights were taken initially, finally, and usually before and after regrounding by means of portable truck scales. Also, since samples were collected at these times, weights of the various constituents could be computed from the analyses and the total gross weights. Care was taken after each windrow or bin content was reground to clean up

TABLE 10.—SUMMARY OF ANALYSES AND WEIGHT CHANGES IN LABORATORY COMPOSTING OF REFUSE.<sup>a</sup>

CONSTITUENT	RAW MIXTURE* PERCENT BY WT.		COMPOST PERCENT BY WT.		SIGNIF. DIFF. AT 95% LEVEL <sup>2</sup>	WEIGHT, LB.		CHANGE IN WEIGHT %	SIGNIF. DIFF. AT 95% LEVEL
	DRY BASIS	VOL. SOL. BASIS	DRY BASIS	VOL. SOL. BASIS		INITIAL	FINAL		
TOTAL WET WT.	100.0 †	—	100.0 †	—	—	40.47 †	27.31	-32.5	YES (-)
MOISTURE	57.1 †	—	58.0 †	—	NO	24.58 †	15.85	-35.5	YES (-)
TOTAL SOLIDS	100.0	—	100.0	—	—	15.89	11.46	-27.9	YES (-)
ASH	7.9	—	12.5	—	YES(+)	1.25	1.43	+14.4	YES (+)
VOLATILE SOLIDS	92.1	100.0	87.5	100.0	YES(-)	14.64	10.03	-31.5	YES (-)
(1) LIPIDS	10.2	11.1	3.3	3.8	YES(-)	1.62	0.38	-76.5	YES (-)
(2) CRUDE FIBER	35.6	38.7	45.9	52.4	YES(+)	5.66	5.26	- 7.1	YES (-)
(3) TOTAL SUGAR	5.0	5.5	0.007 <sup>3</sup>	0.008 <sup>3</sup>	YES(-)	0.80	0.0008	-99.9	YES (-)
(4) STARCH	8.3	9.0	1.3	1.5	YES(-)	1.32	0.15	-88.6	YES (-)
(5) PROTEIN (6.2N)	8.1	8.8	11.0	12.6	YES(+)	1.29	1.26	- 2.4	NO
(6) SUM (1) TO (5)	67.2	73.1	61.5	70.3	YES(-)	10.69	7.05	-34.1	YES (-)
(7) VS-(6) (UNDET)	24.9	26.9	26.0	29.7	NO	3.95	2.98	-24.6	YES (-)
NITROGEN (N)	1.30	1.41	1.76	2.01	YES(+)	0.207	0.202	- 2.4	NO
CARBON (C)	50.1	54.4	47.7	54.5	YES(-)	7.96	5.47	-31.3	YES (-)
RATIO: C/N	38.5		27.1		YES(-)	38.5	27.1	-29.6	YES (-)

\*AS PREPARED FOR COMPOSTING, AFTER SORTING, GRINDING, AND INITIAL WETTING.

† AND ‡ —SEE TABLE 6.

† INCLUDES INITIAL MOISTURE PLUS MOISTURE ADDED DURING COMPOSTING RUNS.

<sup>3</sup> ONLY ONE VALUE (0.2%) WAS ABOVE MINIMUM LIMIT OF ANALYSIS.

<sup>a</sup> AVERAGE VALUES BASED ON 13 RUNS WITH 32 COMPOSTING UNITS (SAVANNAH).  
(MEAN COMPOSTING TIME, 8.8 DAYS, CONTINUOUS AERATION AND MIXING).

as much spilled material as possible, avoiding contamination with soil. Several of the tables show the weights. The weight losses of the various constituents, particularly the dry organic constituents, give perhaps the best indications of degree of decomposition occurring.

Volume was measured in bins, windrows, or trucks. Volume decreased during composting about 36% to 38%. Initial density of the ground, moistened raw refuse was about 630 to 640 lb per cu yd. Final densities were in the range of 610 to 670 lb per cu yd. However, volumes varied widely with several factors, principally degree of compaction and moisture content. Wetting greatly reduces pore space while adding weight, and thereby increases density in

greater proportion than the added weight indicates. Packing of the materials to be composted was avoided as much as possible.

*Sanitary Aspects.*—Close observations of appearance, odor, and fly breeding were made; no nuisances or public health hazards were observed. The composting refuse after only a week had a pleasant brown color and the appearance of finished compost at a distance. While oxygen contents were shown to be quite low in the deeper portions of the windrows, there was rarely any detectable odor near the windrows. On several occasions in windrows and curing piles, a pigsty odor was observed at depths of 24 in. or more. These odors could be detected at distances of 10 ft to 30 ft during regrinding operations, but they disappeared very quickly afterwards. There was rarely any sludge odor from composting of refuse-sludge mixtures even though the raw sludge had become septic in the holding tank prior to initial mixing. Some trouble was encountered in the initial addition of sludge due to run-off into the channel beneath the primary conveyor. This channel was covered by the conveyor belt and was not sloped to drain, so it could not be cleaned out except at the two ends. It would have been better to add sludge to the refuse at the throat of the grinder or in a special mixer following grinding, but circumstances prevented this at the Chandler plant.

On prolonged standing in bins or windrows, the compost developed a dry zone with gray growth on the surfaces. This had a musty or moldy smell, which was not strong or objectionable. Finished compost had an earthy or moldy odor, sometimes scarcely perceptible.

No fly larvae or pupae were observed. Below a depth of 2 in. to 8 in., depending on outer dryness, the composting matter usually was lethal to flies because of its high temperature. After moistening or regrinding, high temperatures approached the outer surfaces. After standing several days, the high temperature progressed inward, but, by then, the outer layer was too dry to be suitable for fly breeding. Thus, the material was either too hot or too dry for fly breeding. Frequent vigorous turning or regrinding would aid in fly control both by killing mechanically and by mixing fly larvae or pupae into hot inner zones.

Flies were observed occasionally on and near the composting materials, generally on cool mornings or after rains or sprinkling. None was observed ovipositing. It is believed that prompt handling of incoming refuse and scrupulous cleaning-up around the plant are very important in fly and rodent control. Beyond that, the high temperatures and frequent turnings of the composting matter should be adequate to control flies. The provision of concrete slabs sloped to drain appears desirable for outdoor composting, and all channels or sumps in the grinding plant should be capable of being washed down and the liquid pumped for use on the compost or to the sewers. As a last resort, the use of chemicals for fly control should take care of emergency situations that might be caused by inclement weather or breakdowns. The use of fly screening at a grinding or a composting plant does not appear indicated except possibly for holding facilities for raw refuse or sludge.

Two other aspects are important: the destruction of pathogenic organisms and of seeds. Both refuse and sludge may contain pathogens. Sludge digestion does not kill all pathogens so it is not much safer to use digested rather than raw sludge. The latter contains more nutrients than the former and, therefore, is more desirable from the standpoint of composting. The temperature curves

(Fig. 1 and 3) indicate that pasteurizing or higher temperatures are maintained for days. In addition, during composting, there is an appreciable effect produced that provides pathogen kills at lower time-temperatures than those lethal to the microorganisms,<sup>9</sup> particularly in the outer or cooler layers where most of the molds occur.

In general, the destruction of pathogens appears to be a statistical problem of determining how many mixings or regrinds are required to insure that all the material is exposed to killing time-temperatures during the processing. In windrow or bin composting, particularly with sludge, it may be necessary to demonstrate complete pathogen destruction before a composting method receives public health approval. Undesirable weed or vegetable seeds should also be killed by the high temperatures. Again, sludge digestion does not destroy these seeds and users of air-dried digested sludge are bothered by the presence of volunteer plants.

### CONCLUSIONS

Although the grossly sorted refuse used at Chandler contained excessive amounts of ash (from glass, metals, ceramics, etc.) and paper, good decomposition was attained in both windrows and aeration bins. More complete sorting for salvage and discard would greatly improve the quality of the refuse, which is believed to be quite similar to that produced at Savannah. Little difference was observed in the decomposition rates in windrows or bins, but use of the former appears to be more practical for a plant installation.

The outdoor bin and windrow composting was conducted in two runs using mixed residential refuse and refuse with raw sewage sludge from Chandler. The grossly sorted and ground materials were turned by regrinding through the plant. Number of regrinds per unit varied from 3 to 8 (average 5.1). Time of composting or composting plus curing varied from 29 to 43 days (average 34.4 days). Decomposition was incomplete but was approaching completion at the end of most runs. About 35% to 40% of the weight of volatile solids (roughly the organic matter) was lost in 30 days of composting and up to 50% in 43 days. A small proportion of nitrogen (average 16.4%) was lost.

In comparison with laboratory mechanical composting, the two outdoor methods appeared to accomplish about the same decomposition in 30 to 40 days as was accomplished in mechanical units with continuous aeration and mixing in about 9 days. In either case, some additional curing may be desirable or necessary. One might therefore expect to decompose good quality refuse or refuse-sludge mixtures in windrows in about 3 weeks with five vigorous turnings plus about 3 additional weeks of curing in sheltered piles or windrows. Similar results might be obtained by composting for about a week in mechanical units, followed by 3 weeks of curing.

Oxygen values were considerably higher in the interiors of the bins than the windrows. About the only apparent benefits from this, however, were less nitrogen losses and a factor of safety in preventing anaerobic odors from being released when the materials were disturbed.

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<sup>9</sup> "Compost Preparation from the Hygienic Viewpoint," by K. H. Knoll, Internatl. Congress on Disp. and Util. of Town Refuse, Scheveningen, mimeo, in English, April 27 - May 1, 1959.

Small cross-sectional area and frequent turning of windrows appear desirable to prevent oxygen depletion and odor production. Frequent vigorous turning or regrinding appears desirable in order to mix, aerate, reduce particle size, and permit moisture adjustment. Turning by simple shoveling or use of a front-end loader is not believed to be adequate. While regrinding may not be practical in a full-scale plant, use of a high-capacity, self-propelled windrow loader should be efficient for windrow turning.

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METABOLISM OF ORGANIC SULFONATES BY ACTIVATED SLUDGE

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SYNOPSIS

The problem of the persistence of synthetic detergents through biological sewage treatment devices and in rivers has been of great concern both in the United States and England. The organic configurations of the commercial alkyl benzene sulfonates that make up the bulk of the syndet washing products on the market today are ones that are resistant to metabolism by bacteria. This study was undertaken in the hope that some information about the mechanism of the metabolism of simple aromatic sulfonates may aid in the continuing research problem of removing syndets from sewage.

Eight activated sludges were developed on four simple aromatic sulfonates and four analog carboxy aromatic compounds. The ability of each of these activated sludges to metabolize the other seven substrates was measured with a Warburg Respirometer. From these data some comments concerning the mechanism of metabolism of the eight substrates could be made.

If the sulfonate radical is the only group on the benzene ring, bacteria will attack at that point, but if any other group is on the ring, this alternate is

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chosen as the point of biological attack. The possibility, therefore, of metabolizing long chain alkyl benzene sulfonates from the sulfonate radical seems remote.

The problem of the persistence of synthetic detergents in treated sewages and in rivers has been one that has plagued sanitary engineers in the United States and England for years. Much research effort has been expended studying the biological metabolism, or lack of metabolism, of commercial alkyl benzene sulfonates. This study was undertaken with the hope that information gained concerning the mechanisms of biological removal of simple aromatic sulfonates by activated sludge might be useful to investigators concerning themselves with this continuing problem of the removal of synthetic detergents from waste waters.

Reports of the lack of complete degradability of the commercial alkyl benzene sulfonate in use in the United States are widespread.<sup>3,4,5,6,7,8</sup> This commercial alkyl benzene sulfonate is a mixture of widely varying structures, long and short alkyl chains, straight and branched alkyl chains, and secondary, and tertiary attachment of the benzene ring to the alkyl side chain. All of these structural variations affect the ease of biological degradability.

D. W. Ryckman, M. ASCE,<sup>6</sup> showed that the attachment of the benzene ring greatly affected the metabolism of alkyl benzene sulfonate. Straight chain alkyl sulfonates of varying chain lengths with a primary ring attachment were readily degraded by activated sludge; similar compounds with a secondary ring attachment were also degraded by activated sludge, although the shorter chain length compounds were more difficult to acclimate. Similar compounds with a tertiary ring attachment were very resistant to biological removal. Branched chain compounds were not studied by Ryckman.

J. F. Nelson, M. ASCE, et al.<sup>9</sup> reported in 1960, on the successful degradation of a straight chain alkyl benzene sulfonate containing a tertiary attachment of the benzene ring. They also studied branched chain alkyl benzene sulfonates and found that the presence of quaternary carbon near the end of the chain prevented successful degradation. Alkyl chains other than straight chains caused reduced metabolism by activated sludge. G. J. Mohanrao<sup>10</sup> has

<sup>3</sup> "Microbiological Degradation of Commercial Alkyl Benzene Sulfonates," by R. E. McKinney, C. N. Sawyer, and J. M. Symons, Progress Report for American Soap and Glycerine Inst. Project, June-September, 1955.

<sup>4</sup> "Bacterial Degradation of ABS, I. Fundamental Biochemistry," by R. E. McKinney and J. M. Symons, Sewage and Industrial Wastes, Vol. 31, 1959, p. 549.

<sup>5</sup> "Bacterial Degradation of ABS, II. Complete Mixing Activated Sludge," by R. E. McKinney and E. J. Donovan, Jr., Sewage and Industrial Wastes, Vol. 31, 1959, p. 690.

<sup>6</sup> "Chemical Structure and Biological Oxidizability of Surfactants," by D. W. Ryckman and C. N. Sawyer, Proceedings, 12th Industrial Waste Conf., Purdue Univ., Lafayette, Ind., 1957, p. 270.

<sup>7</sup> "Effects of Synthetic Detergents on Sewage Treatment Processes," by C. N. Sawyer, Sewage and Industrial Wastes, Vol. 30, 1958, p. 757.

<sup>8</sup> "Removal of ABS by Sewage Treatment Processes," by P. H. McGahey and S. A. Klein, Sewage and Industrial Wastes, Vol. 31, 1959, p. 877.

<sup>9</sup> "The Biodegradability of Alkylbenzene Sulfonates," by J. F. Nelson, R. E. McKinney, J. H. McAteer, and M. S. Konecky, presented at the Amer. Chem. Soc. Meeting at Cleveland, Ohio, on April 12, 1960.

<sup>10</sup> "Activated Sludge Metabolism of Certain Quaternary Compounds," by G. J. Mohanrao, thesis presented to MIT at Cambridge, Mass., in 1959, in partial fulfillment of the requirements for the degree of Doctor of Philosophy. Unpublished.



also shown the ability of activated sludge to metabolize organic molecules containing quaternary carbon atoms. Therefore, as such, the quaternary carbon in some of the alkyl benzene sulfonates in the commercial mixture cannot be considered an absolute metabolic block.

The sulfonate group in an alkyl benzene sulfonate neither presents a problem if the material is degradable (Ryckman<sup>6</sup> showed greater than 90% release of sulfate ion during metabolism), nor seems to be the cause of the lack of degradation in the molecules that are undegradable. The latter statement stems from the agreement of most investigators that the metabolism of alkyl benzene sulfonates begins from the methyl or hydrophobic end of the molecule which dissolves into the lipid portion of the bacterial cell wall. Metabolism then proceeds as far as possible along the alkyl chain, and if complete, into the benzene ring.

R. E. McKinney A. M. ASCE, and E. J. Donovan A. M. ASCE<sup>5</sup> and P. H. McGauhey F. ASCE and S. A. Klein<sup>6</sup> have indicated partial removal of commercial alkyl benzene sulfonate by activated sludge. If this removal were metabolism, it is possible that the degradable molecules in the mixture were being completely metabolized while other structures were untouched. On the other hand, partial metabolism of all of the structures may have occurred, with the resulting end products or fragments diffusing into solution. In either event, the former seeming more likely, sulfonate containing molecules remain in solution. It was the purpose of this investigation to show whether or not sulfonate containing molecules could be attacked from the sulfonate end, thereby bypassing a metabolic block that may exist elsewhere in the molecule.

#### COMPOUNDS STUDIED

The eight compounds studied in this investigation are shown in Fig. 1. Four different simple aromatic sodium sulfonates were studied to show the effect of other groups on the ring. In addition to these sulfonated materials, the analog organic acid to each sulfonate was also studied to show the difference in metabolic patterns of  $R\text{-COONa}$  and  $R\text{-SO}_3\text{Na}$  containing organic compounds.

Reports in the literature concerning the metabolism of simple sulfonated organics are limited. In 1948, J. W. Czekalowski and S. B. Skarzynski<sup>11</sup> tried to grow five different strains of soil bacteria in media containing various benzene derivatives and found no visible growth in the media containing benzene sulfonic acid and p-toluene sulfonic acid. In 1950, N. Kramer and R. Doetsch<sup>12</sup> also had difficulty culturing several species of soil organisms on p-toluene sulfonic acid and benzene-sulfonic acid, but they finally did culture twelve species on benzenesulfonic acid. Subculturing these organisms on sulfosalicylic acid (benzenesulfonic acid with a carboxyl and hydroxy group added) proved successful in only three cases, however.

In 1955, R. H. Bogan A. M. ASCE and C. N. Sawyer<sup>13</sup> successfully developed an activated sludge that would metabolize sodium benzene sulfonate and p-toluene sodium sulfonate. In that same year, McKinney, et al.<sup>3</sup> studied the

11 "The Breakdown of Phenols and Related Compounds," by J. W. Czekalowski and S. B. Skarzynski, *Journal of General Microbiology*, Vol. 2, 1948, p. 231.

12 "The Growth of Phenol Utilizing Bacterial on Aromatic Carbon Sources," by N. Kramer and R. Doetsch, *Archives of Biochemistry*, Vol. 26, 1950, p. 401.

13 "Biochemical Degradation of Synthetic Detergents, II. Studies on the Relation Between Chemical Structure and Biochemical Oxidation," by R. H. Bogan and C. N. Sawyer, *Sewage and Industrial Wastes*, Vol. 27, 1955, p. 917.

metabolism of p-phenol sodium sulfonate, o-monoammonium sulfo sodium benzoate, and p-monopotassium sulfo sodium benzoate by activated sludge. An activated sludge that would metabolize the p-phenol sodium sulfonate could not developed, but sludges were adapted to o-sulfo sodium benzoate in 7 days and p-sulfo sodium benzoate in 28 days. Cross feeding of these two sludges on all three substrates showed a complete absence of simultaneous adaptation.

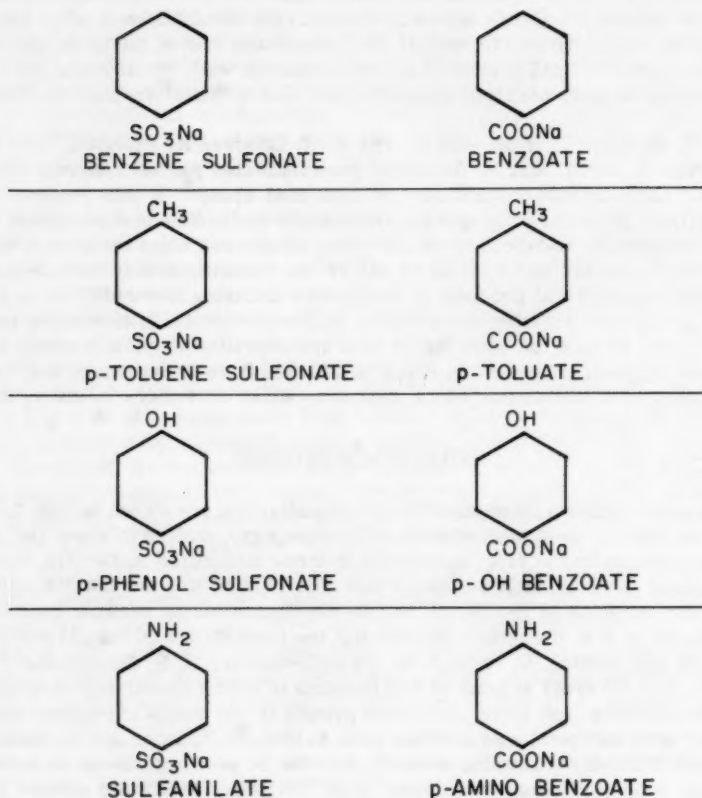


FIG. 1.—COMPOUNDS STUDIED

During this same study an activated sludge developed on phenol was cross fed the three previously noted substrates as well as sodium benzene sulfonate, p-toluene sodium sulfonate, and o-benzaldehyde sodium sulfonate. Of these six substrates, only o-sulfo sodium benzoate was metabolized initially, and that to a limited extent.

In 1957, Ryckman and Sawyer<sup>6</sup> demonstrated the possibility of the metabolism of p-toluene sodium sulfonate and p-ethyl sodium benzene sulfonate by



activated sludge. V. B. Weisbuch<sup>14</sup> in 1959 successfully acclimated an activated sludge to the metabolism of sodium sulfanilate and showed that the amino group and sulfonate group were released into solution simultaneously with the biological removal of COD from solution. In 1960 Malaney<sup>15</sup> reported cross feeding experiments in which an aniline acclimated activated sludge was fed o-, m-, and p-aminobenzene sulfonic acid (the latter compound is sulfanilic acid) and benzene sulfonic acid. Only the m-amino benzene sulfonic acid was metabolized and that only after five days contact in a Warburg Respirometer.

### PROCEDURE

The activated sludges used in this investigation were grown in 1.5 liter, once a day batch fed pilot plants. The operation and control of these systems was essentially the same described by Symons, et al.<sup>16</sup> Each day a quantity of organics, fed as the sodium salt, equal to 1000 mg of COD was added to each system. In addition, sufficient nitrogen, buffer, and Cambridge, Mass., tap water were added to the pilot plants. Initial acclimation was accomplished by adding organics equal to 1000 mg COD and providing continuous aeration until the soluble COD had fallen below 100 mg per l. This cycle was repeated with an ever shortening aeration period, until the system could be fed daily. This daily feeding proceeded for several weeks before the tests were performed on the sludges.

Only one unusual situation developed during the growing of the sludges. The activated sludge grown on p-amino benzoate became quite filamentous after about 7 days to 10 days and would not settle at all. This sludge was discarded, but the same situation developed in the second, and even a third activated sludge grown on p-amino benzoate. Therefore, in order to operate a conventional fill and draw type of pilot plant without losing all of the activated sludge in the effluent, the following procedure was developed: each day 1.0 l of mixed liquor was filtered through coarse fluted filter paper and the solids washed back into the pilot plant with distilled water. Feeding was then accomplished in the usual manner. By this method a high concentration of solids could be maintained in the systems.

Most of the actual testing of the activated sludges was done in the Warburg Respirometer, using 15 ml side arm flasks. The general procedure will be summarized first, followed by an examination of some of the details. A sludge, which had been fed about 24 hr before, was settled, washed twice in distilled water, and finally concentrated to one-fourth of its original volume. This then was a sludge with a mixed liquor suspended solids (MLSS) concentration four times that which existed in the pilot plant (4X sludge). One-half ml of this sludge was placed in the main Warburg flask chamber. To this was added one-half ml of the substrate on which the sludge had been grown, COD equal to 1000 mg per l (IX substrate). Into the side arm was placed 1 ml of another

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14 "The Mechanism of Degradation of Sulfanilic Acid by Activated Sludge," by J. B. Weisbuch, thesis presented to MIT at Cambridge, Mass. in 1959, in partial fulfillment of the requirements for the degree of Bachelor of Science. Unpublished.

15 "Oxidative Abilities of Aniline - Acclimated Activated Sludge," by C. W. Malaney, *Journal of the Water Pollution Control Federation*, Vol. 32, 1960, p. 1300.

16 "A Procedure for Determination of the Biological Treatability of Industrial Wastes," by J. M. Symons, R. E. McKinney, and H. H. Hassis, *Journal of the Water Pollution Control Federation*, Vol. 32, 1960, p. 841.

substrate, COD equal to 2000 mg per l (2X substrate). Eight flasks plus unfed control made up a set. Because the sludge was acclimated to the substrate in the main section of the flask, the oxygen uptake was rapid. When this initial metabolism was finished,  $1\frac{1}{2}$  hr to  $2\frac{1}{2}$  hr, the second substrate was mixed with the sludge and the response of the sludge measured by the resulting uptake. All of the substrates contained organics, buffer, and distilled water only, no nitrogen source.

Simultaneous adaptation was being studied and if the substrates were biologically removed in the pilot plant rapidly, as COD studies showed that they were, the sludges were starved for a long period before the Warburg Respirometer test. During this starvation period adaptive enzymes might be lost, and the sludge might not show the true response to a substrate to which it actually was simultaneously adapted, because the creation of adaptive enzymes by microorganisms is only in response to substrate stimulation. To avoid this possibility use of the side arm flasks allowed each sludge studied to be fed all of the secondary substrates just after the completion of metabolism of its own substrate and when all of the adaptive enzymes, if any, were present in the organism. Studies with and without this preliminary stimulation did not show too much difference in secondary response, but the procedure was continued to cover all possibilities.

The second point is the omission of a nitrogen source from the substrates. Nitrification is one of the most severe limitations on the interpretation of Warburg Respirometer studies. When larger flasks and samples are used, the flask contents may be analyzed for oxidized nitrogen forms when the test is completed and a correction for nitrification made; but this tells nothing of the effect nitrification had on the all important shape of the oxygen uptake curve. Preliminary studies using large flasks and a nitrogen source in the substrate showed that from 17.3% to 30% of the gross oxygen uptake in the fed samples was due to nitrification and from 28% to 55.5% of the oxygen uptake of the unfed control samples was due to nitrification. To reduce these percentages, the sludges were washed in distilled water twice to remove any nitrogen in solution and these washed sludges were fed substrates with no nitrogen source. Of course two of the substrates contained organic nitrogen, so those systems did have nitrogen added. Even in the no nitrogen systems, the possibility of the oxidation of the ammonia released during endogenous respiration still exists, but nitrification was held to a minimum by this procedure.

Because an organism cannot synthesize protoplasm without a nitrogen source, one might think that under these circumstances all of the substrate would be oxidized to carbon dioxide and water and the derived energy wasted as heat. This is not the case, however. As demonstrated by A. F. Gaudy Jr. M. ASCE and R. S. Engelbrecht<sup>17</sup> there is a mass increase during metabolism when nitrogen is absent, and this mass is mostly nitrogen-free polysaccharide. They also showed that the rate of biological removal of an acclimated substrate was relatively unimpaired in an absence of external nitrogen. Data taken in this investigation supports this finding. For example, the initial oxygen uptake rate for the benzoate sludge metabolizing benzoate was only 14% lower when nitrogen was omitted. The other sludges gave similar results.

Gaudy and Engelbrecht<sup>17</sup> also showed that if adaptation is required for the metabolism of a substrate, the lack of an external nitrogen source delays this

<sup>17</sup> "Quantitative and Qualitative Shock Loading of Activated Sludge Systems," by A. F. Gaudy, Jr., and R. S. Engelbrecht, *Journal of the Water Pollution Control Federation*. In Press.

acclimation. This delay is probably due to the inability of the organism to manufacture the required adaptive enzymes because of a lack of protein building blocks. This effect is a benefit to this investigation, because by delaying adaptation due to a nitrogen lack, the separation between the simultaneously adapted substrates and those which require acclimation is wider and more distinct.

## RESULTS

*Sulfonate Sludges Cross Fed Sulfonate Substrates.*—The first series of experiments concerned the response of each of the four activated sludges acclimated to a sulfonate substrate (hereafter called the base substrate) to all four of the sulfonate substrates. The results of this study are shown in Fig. 2. Each of the four sulfonate sludges is shown individually. The first part of the oxygen uptake is associated with the metabolism of the portion of the base substrate that was added to the main part of the Warburg Flask as the test was started. The purpose of this metabolism was to stimulate the sludges. When this first metabolism was essentially finished (time  $1\frac{1}{2}$  or  $2\frac{1}{2}$  hr), the contents of the side arm were dumped into the main part of the Warburg Flask. The occurrence of this second feeding is indicated by a large dot on the oxygen uptake curves. The latter portion of the oxygen uptake curves then shows the response of the stimulated sludge to the secondary substrates. It should be noted that in each case one of the secondary substrates, the dotted curve, is a repeat feeding of the base substrate.

There was some variation in the first or stimulation portion of the oxygen uptake curves for the four replicate sludges used in any one study, but only one average curve is shown to give the general shape and magnitude for all sludges. The variation of the  $1\frac{1}{2}$  hr or  $2\frac{1}{2}$  hr value was usually about 10% from the average.

Except for the p-phenol sulfonate substrate fed the benzene sulfonate [Fig. 2(a)] and p-toluene sulfonate [Fig. 2(b)] sludges, the response to the secondary substrate was clearly rapid and adapted or slow and unacclimated. In the two preceding cases there was moderate metabolism. The secondary portion of the sulfanilate sludge metabolizing its base substrate has been approximately corrected for nitrification. It is interesting to note that the sulfanilate sludge [Fig. 2(d)] showed a typical adaptive shaped oxygen uptake curve both during the stimulatory and second feeding phase of the study. This occurred in spite of the fact that this sludge had been fed daily for over two years before the start of this test. The reason for this daily adaptation probably is connected with the structural similarity between sulfanilate and sulfanilamide. The competition of sulfanilamide with p-amino benzoic acid on the enzyme sites connected with the synthesis of folic acid has been widely reported in the literature and is well summarized by M. J. Pelczar and R. D. Reid.<sup>18</sup> It is quite possible that sulfanilate is somewhat active in the blocking of folic acid synthesis.

In order to help choose the substrates that were adapted, the initial rates of oxygen uptake of the foreign substrates were compared on a percentage basis with the initial rate of oxygen uptake of the base substrate. This latter

<sup>18</sup> "Microbiology," by M. J. Pelczar and R. D. Reid, McGraw-Hill Book Co., Inc., New York, 1958.

TABLE 1

	SUBSTRATE				
	$\text{SO}_3^-$	$\text{CH}_3\text{SO}_3^-$	$\text{OH}\text{SO}_3^-$	$\text{NH}_2\text{SO}_3^-$	
SLUDGE	$\text{SO}_3^-$	100	74	46	22
		100	91	47	23
	$\text{CH}_3\text{SO}_3^-$	106	100	59	27
		116	100	55	24
	$\text{OH}\text{SO}_3^-$	17	8	100	13
		17	13	100	17
$\text{NH}_2\text{SO}_3^-$		68	50	147	100
		16	16	70	100
		29	26	83	100

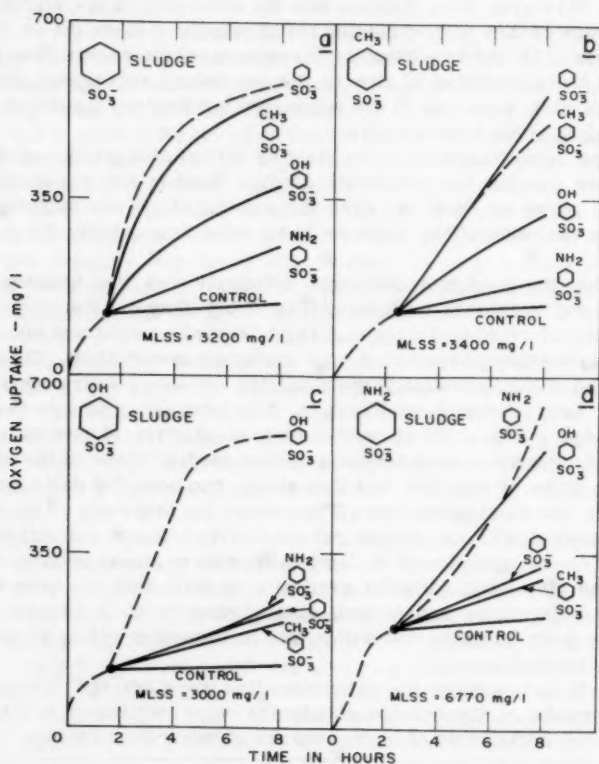


FIG. 2.—RESPONSE OF SULFONATE SLUDGES TO SULFONATE CROSS FEEDING

value was always taken as 100%. In addition, the ratio, as a percentage, of the total oxygen uptake of the foreign substrates divided by this same quantity for the base substrate was calculated. In all cases the total oxygen uptake six hours after adding the secondary substrates was used for the preceding calculation. These data are summarized in Table 1.

Because the sulfanilate substrate fed to the sulfanilate sludge (Fig. 2(d)) showed an adaptive type oxygen uptake curve, a comparison of initial oxygen uptake rates did not seem to show the entire picture. Therefore, the rates of oxygen uptake two hours after the secondary feeding were also calculated as a percentage of the rate of oxygen uptake of the sulfanilate sludge metabolizing its base substrate at time 2 hr. This information is also included in Table 1. These data together with that shown in Fig. 2 were used to select the substrates utilized by the various sludges. These selections are shown in the upper left quarter of Table 5. Only two substrates were indefinite and had to be labeled as questionable.

*Sulfonate Sludges Cross Fed Carboxy Substrates.*—The second series of experiments concerned the response of the four sulfonate activated sludges when the secondary substrates were the carboxy analogs of the four base sulfonate substrates. The results of this study are shown in Fig. 3. In each case the response of a sludge to its base substrate, dotted line, has been included for easy comparison. All of the sludges except the p-toluene sulfonate [Fig. 3(b)] could metabolize their specific carboxy analog and all told seven carboxy substrates were metabolized by the sulfonate sludges.

The percentages of initial oxygen uptake rates and six hour total oxygen uptake are shown in Table 2. As before, additional percentages based on the two hour oxygen uptake rates were calculated for the sulfanilate sludge. The substrates shown in the upper right hand quarter of Table 5 were chosen from the data given in Fig. 3 and Table 2 as being metabolized.

*Carboxy Sludges Cross Fed Carboxy Substrates.*—Similar experiments to those previously described were performed using four activated sludges grown on base substrates that were the carboxy analogs of the sulfonate substrates studied in the first portion of this investigation. The results of this study are shown in Fig. 4 and Table 3. The only surprising aspect of these results is the limited response of the p-amino benzoate sludge [Fig. 4(d)] to its base substrate. This limited response coupled with the continual development of a filamentous activated sludge in the pilot plant indicates that unusual enzyme systems must have to be developed to degrade this compound. The data shown in Fig. 4 and Table 3 were used to choose the substrates placed in the lower right hand quarter of Table 5. Nine substrates were metabolized by the carboxy sludges during this cross feeding.

*Carboxy Sludges Cross Fed Sulfonate Substrates.*—The final phase of this investigation was to determine whether or not any of the activated sludges developed on carboxy substrates could metabolize any of the sulfonate substrates. The results of this study are shown in Fig. 5 and Table 4. In each case the response to the base substrate has been included for easy comparison. Very few of the sulfonate substrates were metabolized by the carboxy sludges. None of the specific analogs was metabolized by the carboxy activated sludges, but benzene sulfonate and p-toluene sulfonate were metabolized fairly well by the p-amino benzoate sludge [Fig. 5(d)]. The results noted in Fig. 5 and Table 4 are summarized in the lower left-hand corner of Table 5.

The substrates shown in Table 5 are those to which the various sludges were either fully adapted (that is, showed an initial oxygen uptake rate equal

TABLE 2

		SUBSTRATE				
		$\text{CO}_2$	$\text{CH}_3\text{CO}_2$	$\text{OHCO}_2$	$\text{NH}_2\text{CO}_2$	
SLUDGE	$\text{SO}_3$	64	51	41	33	% Initial $\text{O}_2$ Uptake Rate
	$\text{SO}_3$	100	53	108	42	% Base $\text{O}_2$ Uptake at 6 hrs.
	$\text{CH}_3\text{SO}_3$	40	26	48	77	% Initial $\text{O}_2$ Uptake Rate
	$\text{SO}_3$	65	26	102	113	% Base $\text{O}_2$ Uptake at 6 hrs.
	$\text{OH}\text{SO}_3$	10	10	83	25	% Initial $\text{O}_2$ Uptake Rate
	$\text{SO}_3$	35	18	89	18	% Base $\text{O}_2$ Uptake at 6 hrs.
	$\text{NH}_2\text{SO}_3$	63	68	135	140	% Initial $\text{O}_2$ Uptake Rate
	$\text{SO}_3$	20	20	63	54	% 2-hour $\text{O}_2$ Uptake Rate
	$\text{SO}_3$	41	22	77	65	% Base $\text{O}_2$ Uptake at 6 hrs.

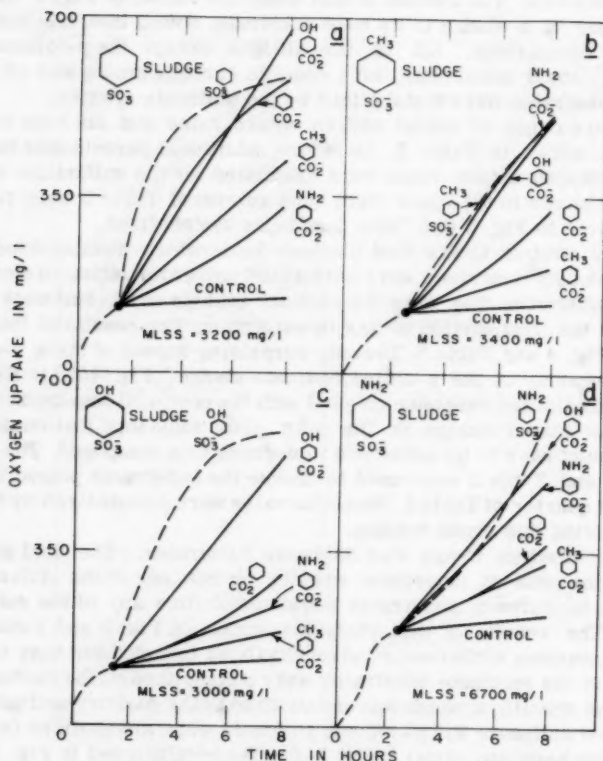










FIG. 3.—RESPONSE OF SULFONATE SLUDGES TO CARBOXY CROSS FEEDING



TABLE 3.—RESPONSE OF CARBOXY SLUDGES TO CARBOXY CROSS FEEDING

		SUBSTRATE				
		 CO <sub>2</sub>	 CO <sub>2</sub>	 CO <sub>2</sub>	 CO <sub>2</sub>	
SLUDGE	 CO <sub>2</sub>	100	19	110	11	% Initial O <sub>2</sub> Uptake Rate
		100	65	109	42	% Base O <sub>2</sub> Uptake at 6 hrs.
	 CO <sub>2</sub>	182	100	60	32	% Initial O <sub>2</sub> Uptake Rate
		102	100	101	46	% Base O <sub>2</sub> Uptake at 6 hrs.
	 CO <sub>2</sub>	77	8	100	8	% Initial O <sub>2</sub> Uptake Rate
		84	50	100	59	% Base O <sub>2</sub> Uptake at 6 hrs.
	 CO <sub>2</sub>	350	64	450	100	% Initial O <sub>2</sub> Uptake Rate
		93	41	117	100	% Base O <sub>2</sub> Uptake at 6 hrs.

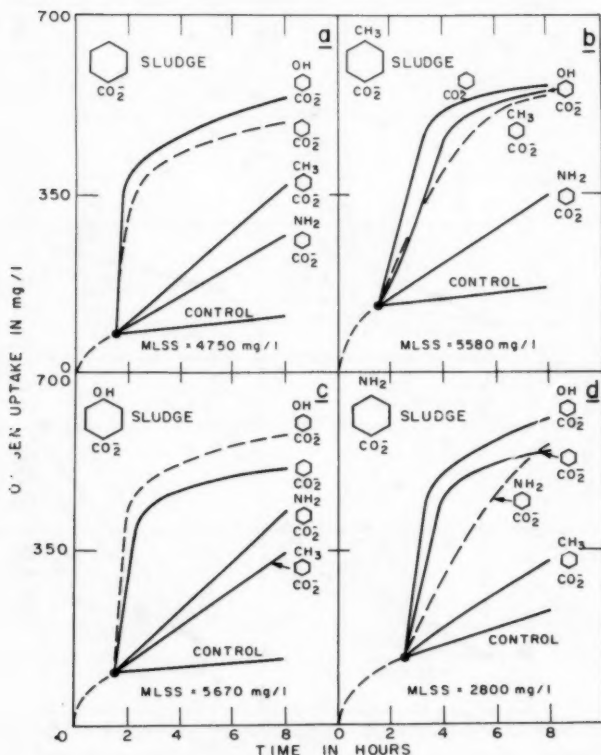


FIG. 4.—RESPONSE OF CARBOXY SLUDGES TO CARBOXY CROSS FEEDING

TABLE 4.—RESPONSE OF CARBOXY SLUDGES TO SULFONATE CROSS FEEDING

	SUBSTRATE				
	$\text{SO}_3^-$	$\text{CH}_3\text{SO}_3^-$	$\text{OH}\text{SO}_3^-$	$\text{NH}_2\text{SO}_3^-$	
$\text{CO}_2$	8	7	9	8	% Initial $\text{O}_2$ Uptake Rate
	26	26	30	30	% Base $\text{O}_2$ Uptake at 6 hrs.
$\text{CH}_3\text{CO}_2$	18	14	19	12	% Initial $\text{O}_2$ Uptake Rate
	19	17	25	17	% Base $\text{O}_2$ Uptake at 6 hrs.
$\text{OH}\text{CO}_2$	7	5	7	5	% Initial $\text{O}_2$ Uptake Rate
	39	37	27	33	% Base $\text{O}_2$ Uptake at 6 hrs.
$\text{NH}_2\text{CO}_2$	83	70	58	48	% Initial $\text{O}_2$ Uptake Rate
	91	72	47	38	% Base $\text{O}_2$ Uptake at 6 hrs.

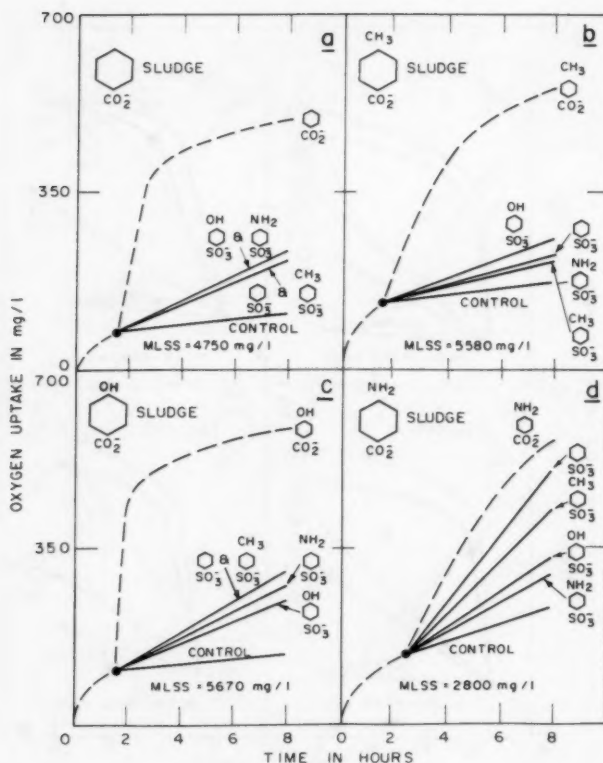






































FIG. 5.—RESPONSE OF CARBOXY SLUDGES TO SULFONATE CROSS FEEDING



to or greater than the base substrate) or showed fairly high oxygen uptake rates. Many of these substrates produced a limited or straight line type of oxygen uptake curve and, therefore, were not fully adapted in the usual sense of simultaneous adaptation. Table 6 summarizes the substrates that yielded typical fully adapted oxygen uptake rates when fed to the various sludges.

TABLE 5.—SUMMARY OF CROSS FEED DATA

		SULFONATE SUBSTRATES METABOLIZED			CARBOXY SUBSTRATES METABOLIZED		
SULFONATE SLUDGES	 SO <sub>3</sub> <sup>-</sup>	 SO <sub>3</sub> <sup>-</sup>	 SO <sub>3</sub> <sup>-</sup>	 SO <sub>3</sub> <sup>-</sup>	 CO <sub>2</sub>	 CO <sub>2</sub>	
	 SO <sub>3</sub> <sup>-</sup>	 SO <sub>3</sub> <sup>-</sup>	 SO <sub>3</sub> <sup>-</sup>	 SO <sub>3</sub> <sup>-</sup>		 CO <sub>2</sub>	 CO <sub>2</sub>
	 SO <sub>3</sub> <sup>-</sup>			 SO <sub>3</sub> <sup>-</sup>		 CO <sub>2</sub>	
	 SO <sub>3</sub> <sup>-</sup>			 SO <sub>3</sub> <sup>-</sup>	 SO <sub>3</sub> <sup>-</sup>	 CO <sub>2</sub>	 CO <sub>2</sub>
	 CO <sub>2</sub>				 CO <sub>2</sub>	 CO <sub>2</sub>	
CARBOXY SLUDGES	 CO <sub>2</sub>				 CO <sub>2</sub>	 CO <sub>2</sub>	 CO <sub>2</sub>
	 CO <sub>2</sub>				 CO <sub>2</sub>	 CO <sub>2</sub>	
	 CO <sub>2</sub>	 SO <sub>3</sub> <sup>-</sup>	 SO <sub>3</sub> <sup>-</sup>		 CO <sub>2</sub>	 CO <sub>2</sub>	 CO <sub>2</sub>
		3	3	4	1	5	1
						8	3

Only fourteen of the sixty-four possibilities were fully acclimated and of those fourteen only five were sulfonate substrates. This shows the general difficulty of metabolism of the simple sulfonates studied.

In Table 6 one of the sludge-substrates combinations, p-toluene sulfonate sludge metabolizing benzenesulfonate, is listed as questionable with respect

to full adaptation. The reason for this is seen in Fig. 2(b). The oxygen uptake curves for both p-toluene sulfonate and benzene sulfonate are very close together; the only difference is that the response to benzene sulfonate was linear, indicating a limited reaction whereas the response to p-toluene sulfonate gave a somewhat typical adapted shape oxygen uptake curve. On the

TABLE 6.—SUMMARY OF FULLY ADAPTED SUBSTRATES

	FULLY ADAPTED SULFONATE SUBSTRATES	FULLY ADAPTED CARBOXY SUBSTRATES
SULFONATE SLUDGES	$\text{C}_6\text{H}_5\text{SO}_3^-$	$\text{C}_6\text{H}_5\text{SO}_3^-$
	$\text{CH}_3\text{C}_6\text{H}_4\text{SO}_3^-$	$\text{CH}_3\text{C}_6\text{H}_4\text{SO}_3^-$
	$\text{OH-C}_6\text{H}_4\text{SO}_3^-$	$\text{OH-C}_6\text{H}_4\text{SO}_3^-$
	$\text{NH}_2\text{C}_6\text{H}_4\text{SO}_3^-$	
CARBOXY SLUDGES	$\text{C}_6\text{H}_5\text{CO}_2^-$	$\text{C}_6\text{H}_5\text{CO}_2^-$
	$\text{CH}_3\text{C}_6\text{H}_4\text{CO}_2^-$	$\text{CH}_3\text{C}_6\text{H}_4\text{CO}_2^-$
	$\text{OH-C}_6\text{H}_4\text{CO}_2^-$	$\text{OH-C}_6\text{H}_4\text{CO}_2^-$
	$\text{NH}_2\text{C}_6\text{H}_4\text{CO}_2^-$	$\text{NH}_2\text{C}_6\text{H}_4\text{CO}_2^-$
	2 2 1 0	4 1 4 0

other hand, the response of the p-toluene sulfonate sludge to its base substrate was not too rapid. This response was in conflict with data taken in the pilot plant itself that showed the COD to be entirely removed from solution and the sulfate ion released into solution three hours after feeding. This is much faster than the data in Fig. 2(b) would indicate. It is possible that the higher

concentration of organics at time "O" in the Warburg Flask, 1000 mg per l COD, as opposed to 667 mg per l COD at the start of each feeding in the pilot plant, could have adversely affected the microorganisms. Other Warburg Respirometer data, however, showed this sludge to exhibit more than twice the initial oxygen uptake rate when fed its base substrate as is shown in Fig. 2(b). This latter statement is in agreement with the soluble COD removal and sulfate release information. For these reasons the possibility of the p-toluene sulfonate sludge being fully adapted to benzene sulfonate is doubtful.

### ANALYSIS OF RESULTS

*General Examination.*—The easiest substrate to metabolize was p-hydroxy benzoate as all sludges were able to metabolize it fairly well and four sludges were fully adapted. The presence of the hydroxy group as an easy point of biological attack on this molecule is the reason for this wide range of adaptability. Benzoate gave a fully adapted response for the four carboxy sludges, but the lack of the hydroxy group prevented any other sludge except the benzene sulfonate sludge from metabolizing this compound, and this in a somewhat limited manner.

Two of the substrates studied were able to be metabolized by only one sludge, the sludge that was developed on the substrate. These were sulfanilate and p-toluate. Of these two, sulfanilate was probably the more difficult to degrade due to its bacterialstatic properties. The reason p-toluate could only be metabolized by its own sludge may have been related to a solubility problem. This compound was much more insoluble than any of the other seven substrates studied; this factor, with the corresponding problem of passage across the bacterial cell wall, surely contributed to the difficulty of metabolism.

Activated sludges developed on sulfonate substrates were able to metabolize seven carboxy substrates on cross feeding, but the carboxy sludges were able to metabolize only two sulfonate substrates. This indicates the inherent difficulty that a sulfonate group on a molecule presents for metabolism. A carboxy group on the other hand is an easy point of biological attack. This fact is also shown in Table 6 in which, although no sulfonate sludge was fully adapted to a carboxy substrate and no carboxy sludge was fully acclimated to a sulfonate substrate, a maximum of five sulfonate substrates were fully adapted while nine carboxy substrates were fully adapted. The size, configuration, and charge on the sulfonate group requires the synthesis of complex adaptive enzymes to allow metabolism.

In general, the more difficult the base substrate is to degrade, the wider the scope of substrates that the sludge was able to metabolize, at least to some degree. The summary numbers in each of the sludge symbols in Table 5 indicate the number of substrates the sludge could handle. The sulfonate sludges could metabolize more substrates than the carboxy sludges, 16 vs 12, and the activated sludge developed on the easiest sulfonate substrate, p-phenol sulfonate, could metabolize only one foreign substrate. The activated sludge developed on the hardest carboxy substrate, p-amino benzoate, ignoring the solubility problem of p-toluate, was able to metabolize the most foreign substrates, including two sulfonate substrates. The necessity for the synthesis of adaptive enzymes for metabolism of the base substrate improves the ability of the microorganism for general adaption. This concept may not be generalized for

all cases, however, and may be valid here only because of the general similarity of all the compounds studied.

The relative ease of metabolism of the eight compounds studied is shown in Table 7. In each case the initial rate of metabolism of the sulfonate sludge is slower than its carboxy analog. Also the first two compounds in each group are more easily degraded than the last two, showing the importance of structure in a general class of compounds.

*Metabolism of Sulfonate Substrates by Acclimated Sludges.*

**Benzene Sulfonate Sludge.**—The only point of attack for benzene sulfonate is through the sulfonate group, unless the aromatic ring is to be split directly as occurs during the metabolism of benzene. The former seems more likely in this case, due to the rapid metabolism of p-toluene sulfonate which may also be attacked through the sulfonate group by an enzyme system adapted to benzene sulfonate; p-Toluene sulfonate might be degraded by oxidizing the methyl group first, but this is not an easy reaction and probably could not occur rapidly in an unacclimated sludge.

The lack of full adaptation to the p-phenol sulfonate indicates that if any other point of attack is available on a sulfonate containing molecule, it will be utilized in preference to the sulfonate group. In this case, in spite of an enzyme

TABLE 7.—INITIAL RATE OF OXYGEN UPTAKE OF SLUDGES METABOLIZING THEIR BASE SUBSTRATES

Sulfonate Sludges	Rate, in mg per l per hr	Carboxy Sludges	Rate, in mg per l per hr
Benzene Sulfonate	133	p-Hydroxy benzoate	550
p-Phenol sulfonate	120	Benzoate	310
p-Toluene sulfonate	70	p-Toluate	156
Sulfanilate	58	p-Amino benzoate	110

system that could metabolize the sulfonate group, p-phenol sulfonate was attacked through the easier hydroxy group even though this required some acclimation. The sulfanilate substrate was not utilized, probably due to its bacterial-static properties.

**p-Toluene Sulfonate Sludge.**—Due to the doubtfulness of the full adaptation of this sludge to benzene sulfonate, the point of attack of p-toluene sulfonate is in question. Both groups of this molecule are difficult to attack initially, the question is which is more difficult, the methyl or sulfonate group? If benzene sulfonate is fully adapted, then p-toluene sulfonate is attacked through the sulfonate group. If in Fig. 2(b) the initial rate of oxygen uptake of the p-toluene sulfonate substrate was slower than usual for some reason, and the response to benzene sulfonate was correct, then p-toluene sulfonate was metabolized through the methyl group. Based on other oxygen uptake data, the rate of COD removal from solution in the pilot plant, and the rate at which benzene sulfonate can be metabolized by a well acclimated sludge [Fig. 2(a)], about twice the rate at which the p-toluene sulfonate sludge metabolized it) the writers feel that the

methyl group is the point of attack. The other two sulfonate substrates, studied were not fully adapted for the same reasons previously outlined.

**p-Phenol Sulfonate Sludge.**—p-Phenol sulfonate is metabolized through the hydroxy group, as indicated by the inability of this sludge to metabolize any of the other sulfonate substrates. The presence of a ready point of attack made this compound the easiest of the substituted sulfonate substrates to metabolize.

**Sulfanilate Sludge.**—Sulfanilate was hard to metabolize due to its structural similarity to sulfanilamide, but when it was metabolized it was attacked at the amino group end. This is shown by the lack of full adaptation to benzene sulfonate. p-Phenol sulfonate could be metabolized by this sludge due to the ready point of attack at the hydroxy group, while the hard to metabolize methyl group on p-toluene sulfonate probably prevented its utilization.

### SUMMARY

From the preceding examination it would appear that, although it is possible to degrade a sulfonate molecule from the sulfonate end, if there is any other group present on the aromatic ring, this alternate group is the point chosen for attack. This is true even if it is difficult to metabolize the alternate group itself; for example, the methyl group of p-toluene sulfonate. This would indicate that the possibility of degrading a large alkyl benzene sulfonate molecule from the sulfonate end is remote. This is particularly true of a synthetic detergent, because the sulfonate end is hydrophilic, and so the methyl end enters the microorganism first. But even considering fragments of syndet as remnants of partial syndet metabolism, it seems unlikely that an activated sludge could be stimulated into metabolizing these fragments from the sulfonate end. The large size, configuration, and high negative charge makes the sulfonate group unappealing to most microorganisms.

### CONCLUSIONS

From this investigation the following conclusions with respect to these eight compounds may be drawn:

1. The presence of a sulfonate group makes a compound hard to degrade.
2. Even with an easy point of attack such as a hydroxy group on a sulfonate molecule, an unacclimated activated sludge cannot readily metabolize the molecule.
3. Activated sludges acclimated to sulfonate substrates can use carboxy substrates fairly readily, but the reverse is not true.
4. If any other point of attack is available on a sulfonate molecule, it will be utilized in preference to the sulfonate group.
5. The ease of metabolism of a substituted sulfonate compound is generally related to the ease of attack on the substituted group, but spacial configuration also has an affect.
6. Benzene sulfonate is metabolized through the sulfonate group.
7. p-Toluene sulfonate is probably metabolized through the methyl group.
8. p-Phenol sulfonate is metabolized through the hydroxy group.
9. Sulfanilate showed bacterialstatic properties, but was metabolized through the amino group.

10. As indicated by the p-amino benzoate metabolism, the presence of an amino group in an aromatic compound does not provide too easy a point of biological attack.

#### ACKNOWLEDGEMENT

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Journal of the  
SANITARY ENGINEERING DIVISION  
Proceedings of the American Society of Civil Engineers

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DISCUSSION

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SOLUBILITY OF ATMOSPHERIC OXYGEN IN WATER<sup>a</sup>

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Closure by the Committee on Sanitary Engineering Research  
of the Sanitary Engineering Division

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Since publication of the original paper in July 1960, substantial activity regarding the subject of dissolved oxygen saturation has taken place. I. C. Morris, W. Stumm, M. ASCE and H. A. Galal describe the details of their experiments and raise several questions regarding the experimental conditions used in the report. I. C. Hart and Downing report a careful reappraisal of the analytical techniques used by G. A. Truesdale, A. L. Downing, and G. F. Lowden,<sup>22</sup> and indicate a strong possibility that the volatilization of iodine in the Winkler titration used in their laboratory was the reason for their values being lower than those given in the report and found in other experimenters. The newly published eleventh edition of *Standard Methods*<sup>23</sup> omits the second decimal place in the G. C. Whipple and M. C. Whipple<sup>24</sup> computation of the C. J. J. Fox<sup>25</sup> saturation values, thus implying a reduction of confidence in these values. B. B. Benson and P. O. M. Parker<sup>26</sup> have completed a study of the solubility of argon, nitrogen, and oxygen in water and used a unique scheme of interconsistent ratios to conclude that the values reported in the report are the most reliable for oxygen. J. Carpenter at The Johns Hopkins University<sup>27</sup> has under way an extensive program of experimentation that should be quite helpful when completed.

The prior mentioned discussions from the Harvard group and from the Water Pollution Research Laboratory are welcomed, inasmuch as they emphasize the interest and the practical importance of accurate dissolved oxygen saturation values.

The points of concern expressed by Morris, Stumm, and Galal are related to (1) barometric pressure measurement, (2) relative humidity, and (3) the technique of computation used. It is thought the following information will illuminate each of these subjects.

(1) The rudimentary correction of the barometric pressure to standard conditions was conducted before the data were processed. Standard corrections were included for the relative differences in thermal expansion of the

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<sup>a</sup> July 1960, Report of the Committee on Sanitary Engineering Research of the Sanitary Engineering Division (Proc. Paper 2556).

<sup>22</sup> G. A. Truesdale, A. L. Downing, and G. F. Lowden, *Journal of Applied Chemistry*, Vol. 5, 1955, p. 53.

<sup>23</sup> "Standard Methods for the Examination of Water and Wastewater," APHA, New York, 11th Ed., 1960.

<sup>24</sup> G. C. Whipple, M. C. Whipple, *Journal of American Chemical Society*, Vol. 33, 1911, p. 362.

<sup>25</sup> C. J. J. Fox, *Transactions, Faraday Soc.*, Vol. 5, 1909, p. 68.

<sup>26</sup> B. B. Benson and P. D. M. Parker, *Journal Physical Chemistry*.

<sup>27</sup> J. Carpenter, Private Communications, June 27, 1960.

brass case and the glass tube, as well as the latitude-gravity factor and the altitude-gravity factor. The barometer (as well as the barograph) was located within the same laboratory room that contained the thermostated water bath. Special care was used to detect variations in barometric pressure between the interior of the building and the outside atmosphere, but none was found.

(2) Early experimental equipment included a wetted-wall column to insure that the air entering the reaction vessel would be completely saturated. Comparative tests showed, however, that this device was unnecessary, since results with, and without, were identical within the accuracy of the test. The reaction vessel used was quite large (45 l) and thus the airspace above the water allowed a relatively lengthy period for the entering air to become saturated with water vapor. With a 20-l airspace available, this would produce a theoretical displacement interval of 80 min. A lesser rate of air entry was not thought desirable in that less assurance of the total removal of excess nitrogen or oxygen would result.

(3) In common with the practice of Fox<sup>25</sup> and Truesdale et al.,<sup>22</sup> a cubic equation was selected to provide ample flexibility and thus permit a good fit of the experimental data, and to provide a firm basis for interpolated values. There is no question concerning the goodness-of-fit of the line of regression to the 260 experimentally determined points which were grouped around seven temperatures relatively evenly spaced between 1.8°C and 29.1°C. With a coefficient of correlation of 0.99980 and 3° of freedom, a significance level well beyond the 1% point is indicated. (Any value greater than 0.983 is considered highly significant.)

There is no intention to suggest the use of the specific derived equation to fit the full range of saturation values, and certainly there would not be expected a good, or even an approximate, fit when extrapolated to 100°C. The occurrence of factors that are insignificant within the experimental temperature range, but which assume significant proportions at higher temperatures, precluded such a possibility. For example, the effect of vapor pressure of water on barometric pressure is hardly significant within the 0° to 30° range; but at higher temperatures, this factor assumes considerable importance.

Despite the desirability from a theoretical standpoint of extending the experimental data to as high as 50°C, the 0°C to 30°C range covered in these experiments quite adequately covers the normal temperature range found in natural streams.

The discussion of Hart and Downing is concerned with the analytical aspects of precise dissolved oxygen determination. The analytical technique used in the report was evolved from several years of research covering the various electroanalytical methods that are available. A more detailed account of the procedure is presently being prepared for publication, and a copy will be supplied to the English group for study and appraisal.

It is agreed with Hart and Downing, that the volatilization of iodine is an important variable in dissolved oxygen titrations. In fact, when either a manual or recording polarograph is used as readout in the direct amperometric titration of iodine with thiosulfate, the effect of the disappearance of the iodine can be visually observed on the instrument. Therefore, it is essential that back-titration be used when ultra-precision is required. This is in agreement with the findings of G. Knowles and Lowden.<sup>28</sup> Volatilization of iodine would well account for the differences noted in the values reported by

<sup>28</sup> "Analyst," by G. Knowles and G. F. Lowden, Vol. 78, 1953, p. 159.

the various groups of researchers. The forthrightness of Hart and Downing in admitting the possibility of this defect in the procedure of Truesdale, Downing, and Lowden, is admirable. Studies by Hart and Downing to fully delineate the effect of iodine volatilization will be most helpful to the field.

For future work in this field, three suggestions are proposed:

1. The analytical procedure used must be capable of a high degree of precision, and this precision must be attainable on samples taken in the normal manner from the reaction vessel being used;
2. Experimental conditions should be meticulously controlled, carefully monitored, and recorded; and
3. Extensive replication must be practiced to eliminate the effect on the final results of randomly occurring errors that are an inseparable adjunct of all experimental work.

The contributions to knowledge in this field made by those who prepared discussion of this paper are much appreciated. The interest stimulated in the definitive determination of these widely used values is most gratifying. With so many groups working or interested in this field, there is little doubt that the end result will be the development of a final set of values for dissolved oxygen saturation that will resist the test of time.

#### ACKNOWLEDGMENTS

The report and closure were prepared by Harold L. Elmore, Public Health Engineer, and Theodore W. Hayes, Analytical Chemist, Stream Pollution Control Section, Division of Health and Safety, Tennessee Valley Authority.

The Sanitary Engineering Division gratefully acknowledges the generosity and professional courtesy of the authors of this paper in making the results of their research work available for publication. The SED Research Committee also recognizes the cooperation of the Tennessee Valley Authority in releasing this paper for publication herein.

This research report, which is one of a series of professional contributions by The Committee on Sanitary Engineering Research, has been obtained for publication, reviewed and approved, by the Stream Pollution Section.

M. A. Churchill, F. ASCE Head



LOW PRESSURE AERATION OF WATER AND SEWAGE<sup>a</sup>

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Closure by N. Claes H. Fischerström

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N. CLAES H. FISCHERSTROM,<sup>1</sup> F. ASCE.—Gánczarczyk has given some valuable additional data regarding low pressure aeration of water. His results correspond well to the writer's although his experimental equipment might have been somewhat different to that at Nacka. They are of special interest however, with regard to the results from investigations of a "second" efficiency optimum, which results will strongly support the conclusions in the paper on future developments of the activated sludge process.

Gánczarczyk's general conclusions are in good agreement with the writer's. Fig. 25 and Fig. 26 are similar to Fig. 20 and Fig. 18 though the diagram axis are chosen different. Fig. 25 indicates an increase of the oxygenation capacity in direct proportion to the air rate but for the highest oxygenation capacities. Fig. 26 indicates the most economic immersion depth to be about 800 mm. The writer has found (Fig. 18) a most economical depth of 2.5 ft to 3 ft or 750-900 mm.

The numerical values of the oxygenation also show a good conformity. At 800 mm depth the writer has determined, at an aeration level of 100, an oxygenation capacity for a normal distributor of 700 g per m<sup>3</sup> h (Fig. 20) and Gánczarczyk an oxygenation capacity of 670 to 700 g per m<sup>3</sup> h. At lower aeration levels Gánczarczyk has indicated better oxygenation capacities than the writer, for example 400 instead of 300 g per m<sup>3</sup> h at an aeration level of 50. He has also shown in Figs. 27 and 28 that the higher oxygenation capacity-values mainly depends on a maximum efficiency of the aerator at an aeration level of approximately 50-80. He also points to the corresponding low head losses of the air distributor. Maximum oxygenation is given as 3.3 kg O<sub>2</sub> per  $\phi$ ,  $\phi$  kilowatt hour netto, corresponding to perhaps 2.5 kg O<sub>2</sub> per kWh brutto.

From Fig. 27, with its indicated variations in efficiency at different rates, it can be seen that at conventional oxygenation capacities, aeration levels of about 10, the power consumption is nearly 100% higher than at best efficiency at an aeration level of approximately 60. Because of the conservatism in the sewage field, little of the results of the last 10 yr research has been adopted in the purification plant design, the possibilities and advantages of using a high oxygenation rate and a short detention time in the activated sludge process again should be emphasized.

Gánczarczyk has found that the hydraulics of the water body is irregular and that tests should be conducted in basins of the actual size. This is in close agreement with the writer's conclusions as shown under the headings "Circulation Rates" and "Economic Considerations." The same can be said about

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<sup>a</sup> September 1960, by N. Claes H. Fischerström (Proc. Paper 2607).

<sup>1</sup> Senior Cons. Engr., Vattenbyggnadsbyran, Stockholm 5, Sweden.

the construction of the air distributor; compare the writer's conclusions, point 7.

*Errata.*—Page 22, line 15, change "they" to "there," line 16, "difficult" to "difficulties," delete "of," "material" to "materials"; page 26, Fig. 4 is upside down; page 30, line 3, change " $<0$  at  $h_2 = 0.36$ " to " $<0$  at  $h_2 = 0.36$ ," line 13, 5 to 6; page 31, line 30, delete 2; page 32, line 18, should read "(PH 1.5 to 11)," line 25, change "couble" to "double," line 27, add "Imhoff" after "1953"; page 33, Table 1, lines 9, 10, 11, change "D per o" to "D: o," Table 2, line 8, change "cone" to "conc," lines 10, 11, change "D per o" to "D: o"; page 35, line 10, delete "It is not quite clear if," line 11, add "and the" after "factory," change "valve" to "value," Table 3, lines 11, 12, change "D per  $\phi$ " to "D: o"; page 41, line 18, change " $t, ^\circ\text{C}, K_{10}$ " to " $t^\circ\text{C}, K_{10}$ ," line 28, change " $L = Q_A Q_B$ " to " $L = Q_A : Q_B$ "; page 43, line 2, delete %; page 49, line 1, change "arossion" to "erosion," line 6, change "added" to "is caused," line 45, change "K hr" to "kwhr," line 46, add after "in" "pipes, distributors," line 48, add after "for" "clean filters, distributors, and pipes and for;" page 50, line 4, add after "clogging" "of filters and distributors," line 8, add the sentence, "The factors  $K_1$  and  $K_2$  must be estimated by the advising engineer," lines 11, 12, 13, change first "K hr" to "kwhr"; page 54, Table 4, in Remarks, line 2, change "L P" to "11-14," line 7, change "N to 13"; page 55, line 5, change "is for" to "has been," "and" to "by," line 12, after "not the" add "amount of air," line 15, change "K hr" to "kwhr."



DEVIATION OF FLOW EQUATIONS FOR SEWAGE SLUDGES<sup>a</sup>

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Closure by Vaughn C. Behn

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VAUGHN C. BEHN,<sup>24</sup> F. ASCE.—Bugliarello has made a useful contribution to the original paper which should be of great assistance to those who have a serious interest in the application of non-Newtonian technology within the field of sanitary engineering.

Because many of Bugliarello's remarks are concerned with applications under the conditions of turbulence, it should be mentioned that not all of the field of applications of waste sludge transportation are in the turbulent range. D. R. Tatman, A.M. ASCE, has reviewed a number of cases from the literature and has concluded that American practice tends in this direction, but that English practice (which is at least or more extensive) tends to laminar flow.<sup>25</sup> To make the necessary computations, the non-Newtonian properties must of course be estimated. In an actual application of the procedures suggested by Bugliarello and the writer, the design engineer should not overlook the scale-up methods described by Alves, Boucher, and Pigford.<sup>7</sup>

Bugliarello's comment on the lack of rigor of the pseudoplastic power-law equations when  $K$  and  $n$  are not constant are completely valid, but it is to be hoped that the paper was not misleading on this point. When these equations do not hold a two-parameter system it is evidently not sufficient. The writer concurs in the necessity for stipulating the range of shear stress when these equations are used, and in fact such a procedure has been carefully followed in the presentation of some data on digested sewage sludge.<sup>26</sup>

The additional rigor of the equations for any time-independent non-Newtonian and the restriction against time dependency is believed to be implied in the title. Based primarily on experience with different length capillary tubes, the writer is inclined to believe that the waste sludges encountered to date are sufficiently time-independent to encourage the engineering application of the generalized equations.<sup>27</sup> These equations have formed the basis for most of the analysis of capillary viscometer data collected in our laboratory. The rotational viscometer is another matter. In data collected to date (1961) at the relatively low shear rate with the MacMichael, it has not mattered whether

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<sup>a</sup> November 1960, by Vaughn C. Behn (Proc. Paper 2663).

<sup>24</sup> Assoc. Prof. Civ. Engrg., Cornell Univ., Ithaca, N. Y.

<sup>25</sup> Thesis, by D. R. Tatman, presented to the Univ. of Delaware, at Newark, in June, 1961, as partial fulfillment for the requirements of the degree of Master of Science in Civil Engineering.

<sup>26</sup> "Application of the Method of Composition and Linear Regression Analysis to a Problem Involving a Pseudoplastic Suspension," by W. F. Ames and V. C. Behn, Transactions, Rheological Soc., 1961.

<sup>27</sup> Thesis by J. Sirman, presented to the Univ. of Delaware, at Newark, June, 1960, as a partial fulfillment of the requirements for the degree of Bachelor of Science in Civil Engineering.

the generalized equations in the power-law equations were used. This is because behavior has been power-law and the term involving the derivative of  $n'$  (Eq. 161 and Appendix III) has been negligible.

Concerning the use of rotational viscometers generally some difficulty has been experienced in utilizing the equipment for heavy sludges. Also, difficulty has been experienced in obtaining independent flow diagrams which would coincide for capillary derived diagrams using the same fluid. This same uncertainty concerning the instrument is reflected in Bugliarello's comments on the onset of turbulence in the rotational viscometer.

Bugliarello suggests the alternative consideration of the plate and cone type viscometer. As of this time no attempt has been made to procure one of these for our laboratory work. The reasons for this are not binding and are simply a matter of a relatively higher cost and the concern about their application to suspensions.

Finally the writer would like to record the informal comments of Peter Monkmeyer, A.M. ASCE, of the Cornell Staff. The conversion to the basic shear stress-shear rate relationship in viscometers (Eq. 88) is accomplished more rigorously via cylindrical coordinates.<sup>28</sup>

*Errata.*—The following equations should be corrected to the form given.

$$\int_{v_c}^{v_w} dv = - \int_{r_c}^{r_w} \frac{\tau_w r}{\mu r_w} dr \dots\dots\dots (23)$$

$$v_c = \frac{\tau_w D}{4 \mu} \dots\dots\dots (24)$$

$$v_i = \frac{\left(\frac{\tau_w}{K}\right)^{\frac{1}{n}} r_w}{(1 + 1/n)} \left[ 1 - \left(\frac{r_i}{r_w}\right)^{(1+1/n)} \right] \dots\dots\dots (46)$$

$$\int_0^{r_w} \left[ \int_{r_i}^{r_w} \frac{r dr}{\mu_a} \right] r_i dr_i = \left[ \frac{r_i^2}{2} \int_{r_i}^{r_w} \frac{r dr}{\mu_a} \right]_{r_i=0}^{r_w} - \int_0^{r_w} \frac{r_i^3 dr_i}{2 \mu_a} \dots\dots (59)$$

$$\int_0^{r_w} \left[ \int_{r_i}^{r_w} \frac{r dr}{\mu_a} \right] r_i dr_i = 0 + \int_0^{r_w} \frac{r_i^3 dr_i}{2 \mu_a} = \frac{1}{2} \int_0^{r_w} \frac{r^3 dr}{\mu_a} \dots\dots (60)$$

$$\frac{dx}{d\tau_w} \tau_w^3 + 3 \tau_w^2 x = \frac{\tau_w^3}{(\mu_a)_w} \dots\dots\dots (66)$$

$$\Omega = \frac{G(\tau_1) \ln S}{\left[ 1 + m \ln S + \frac{1}{3} m^2 (\ln S)^2 + \frac{(\ln S)^2}{3} \frac{dm}{d(\ln \tau_1)} + \dots \right]} \dots\dots (115)$$

<sup>28</sup> "Boundary Layer Theory," by Herman Schlichting, Fourth Ed., McGraw Hill Book Co., 1960, p. 54.

$$\frac{\Delta P}{L} = K_3 \left( \frac{\rho V^2}{D} \right) \left( \frac{V D \rho}{\eta} \right)^{h-2} \left( \frac{D^2 \rho \tau_y}{\eta^2} \right)^b \dots\dots\dots (121)$$

$$f = K_3 (N_{RE})^{h-2} (N_{HE})^b \dots\dots\dots (122)$$

$$N_{RE} = \frac{8 V^{2-n} D^n \rho}{K \left[ \frac{6n+2}{n} \right]} \dots\dots\dots (127)$$

$$\left( \frac{1}{S^2} \right)^m = 1 + m \ln \left( \frac{1}{S^2} \right) + \frac{m^2 \left( \ln \frac{1}{S^2} \right)^2}{2!} + \frac{m^3 \left( \ln \frac{1}{S^2} \right)^3}{2!} + \dots\dots\dots (149)$$

On page 74, line 10, change  $(N_{RE})^{n-2}$  to  $(N_{RE})^{h-2}$



CHLORINATION OF MIXED SEWAGE AND STORM WATER<sup>a</sup>

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Closure by Thomas R. Camp

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THOMAS R. CAMP,<sup>13</sup> F. ASCE.—The writer is grateful for the discussion contributed by P. E. Seuffer relative to the waste of taxpayers' money which results from arbitrary requirements of more treatment of sewage than is actually needed to comply with the stream water quality standards. This is a problem of grave importance to the Nation and it must be made a matter of increasing concern to all sanitary engineers. We must clean up our streams, but must not go hog-wild in the construction of a large number of unnecessary sewage treatment plants. Stream standards must be met, but any expenditure in excess of the minimum required for this purpose is economic waste.

The principal purpose of the paper was to point out that despite the hue and cry by Federal and State officials for more and more sewage treatment plants to save our water resources, practically no attention is being paid by these same officials to the discharge of mixed raw sewage and storm water into our water courses every time it rains. This constitutes gross pollution during rainstorms which in many cases exceeds the pollution allowed by the stream standards. In the writer's opinion the pathogenic bacteria and viruses accompanying these discharges are of far more significance to the public in most cases than the removal from the sewage of suspended solids and BOD. The most important purpose of sewage treatment should be to destroy pathogenic bacteria and viruses.

The second purpose of the paper was to demonstrate that screening and chlorination of mixed sewage and storm water overflows is feasible and is not as expensive as might be inferred from the high ratio of the rate of flow of storm water to the rate of flow of sanitary sewage. The writer's studies indicate that at the locations considered the total amount of storm water which must be chlorinated throughout the year is only about 40% of the dry-weather flow of sanitary sewage. The writer concluded that the total chlorine required for chlorinating the storm water and the dry-weather flow of sanitary sewage would be only about 20% greater than the chlorine required for the dry-weather flow. This conclusion was based primarily on the results of the experiments conducted by McCarthy at the Lawrence Experiment Station in which sterile tap water was used to simulate the storm water which would be mixed with the sewage in overflows. Since the presentation of the writer's paper in October, 1960, it has been brought to his attention by several persons that this conclusion is faulty on two counts.

*Chlorine Demand Higher During Storms.*—The first count is that it cannot be assumed that the suspended matter and bacteria in sewage will be diluted

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<sup>a</sup> January 1961, by Thomas R. Camp (Proc. Paper 2701).

<sup>13</sup> Camp, Dresser & McKee, Cons. Engrs., Boston, Mass.

by storm water in proportion to the amount of storm water present. The reason for this is that the velocities in combined sewers during dry weather are so slow that a considerable amount of suspended matter settles on the bottoms of the sewers. This accumulation is greater the longer the period of dry weather between storms. When rainstorms occur the velocities increase considerably and much of the deposited suspended matter is picked up and mixed with the sewage and storm water. The writer has been informed that some analyses have indicated that the chlorine demand of mixed sewage and storm water even during high storm flows may be as high as or higher than the chlorine demand of the dry-weather sewage and that the chlorine demand of the mixed sewage and storm water may remain high for considerable periods after the beginning of the storm. This information is correct and there is considerable evidence to substantiate it. It will be shown later that this information has tremendous significance in cases where the receiving waters require the removal of suspended solids and BOD.

If a combined sewer has a full capacity of 50 to 100 times the average dry-weather flow, the depth of flow during dry weather will be only about 10% of the diameter of the sewer if it is circular and less if it has a relatively flat bottom. The velocity during dry weather will average about 30% of the full velocity. With a full velocity of 3 fps, the average velocity during dry weather will be only about 0.9 fps. In order to maintain an average velocity in excess of about 1.6 fps for adequate cleansing, the full velocity would have to exceed 5.4 fps.

In studies of the chlorine demand of the Buffalo sewage during the years 1938 to 1940 Symons<sup>14</sup> et al found that on 126 days when rain occurred, the average chlorine demand before the storm was 5.3 ppm, after the first hour was 6.0 ppm, after the second hour was 6.8 ppm and diminished slowly to 5.8 ppm after 6 hr. These were the average values for 126 days of storm. In cases where rains continued from 8 hr to 24 hr, the demand was usually less after the first few hours. In cases of sudden sharp showers the demand usually increased greatly during the first hour and returned to normal within an hour or two. The Symons data confirm the claims that with most combined sewer systems high chlorine doses must be used even with high dilutions of storm water to sewage.

It appears from the preceding that it may be impractical to apply chlorine in sufficiently concentrated doses to assure a free chlorine residual of 1 ppm for 10 min contact at the design capacity of the chlorination plant as proposed by the writer. This is the requirement indicated by the studies of Kelly and Sanderson for effective virus kills. It may be necessary to forego effective virus kills for all flows great enough to produce a contact period less than about one hour. If the dose is great enough to produce a combined chlorine residual of at least 10 ppm for all flows up to the flow required for a contact period of about 1 hr, effective virus kills should result for these smaller flows according to the studies of Kelly and Sanderson. This would mean sufficient chlorinator capacity for doses of 25 ppm to 30 ppm at flows up to about 1/4 to 1/2 the capacity of the combined sewer. With this chlorinator capacity, the concentration of the dose would decrease at higher flows down to 5 ppm to 10 ppm at the full capacity of the combined sewer and chlorination plant. With

<sup>14</sup> "Variation in the Chlorine Demand of Buffalo Sewage," by G. E. Symons, R. S. Simpson, and S. R. Kin, *Sewage Works Journal*, Vol. XIII, No. 2, March, 1941, p. 249.

this method of design it may be expected that the total annual chlorine requirements for chlorinating the storm water mixed with the sewage and all the sewage would be about 50% greater than the chlorine required for the dry-weather flow, instead of about 20% greater as indicated in the paper.

*No Free Chlorine in Tests at Lawrence.*—The other point brought to the writer's attention by several persons subsequent to October, 1960, relates to the comparative values of the chlorine doses and the concentrations of free ammonia in the mixed sewage and simulated storm water in the McCarthy experiments, Tables 4 to 9, inclusive. It was intended to have a free chlorine residual in these experiments of 1.0 ppm after 20 min contact time. In order to estimate the required chlorine dose it was necessary to determine the "combined chlorine demand" which was defined as the chlorine dose required to yield a free chlorine residual of less than 0.05 ppm after 20 min contact time. The actual dose used was 1.0 ppm in excess of this combined chlorine demand. It will be noted in Tables 4 to 9, inclusive, that the chlorine dose was of the same order of magnitude as the free ammonia nitrogen in the diluted sewage and in some cases was less. It is well known that in water chlorination, a chlorine dose of about five times the content of free ammonia nitrogen by weight is required to convert all the free ammonia to monochloramine and an additional chlorine dose of about the same amount is required to reach the "break point" where the monochloramine has all been converted to dichloramine and the dichloramine has all been decomposed. This anomaly was discovered when the results of the McCarthy tests were at hand just prior to the presentation of the paper, but time was not then available to explore the cause of the discrepancy.

The most likely explanations for the results obtained in the McCarthy tests are that: (1) the analytical procedure used for the determination of free ammonia may have yielded results which were too high, or (2) the analytical procedure used for the determination of free residual chlorine may have yielded high results, or (3) both. Experimental studies have subsequently been made by both McCarthy and by the writer's associate, Robert H. Culver which throw considerable light on this question.

At the suggestion of J. Carrell Morris of Harvard University, Culver processed the Lawrence sewage for free ammonia analysis by centrifuging and then extracting the free ammonia with an ion exchange column. It was found that the free ammonia content of Lawrence sewage was approximately the same when obtained by this method as by the procedures outlined in "Standard Methods," and it was further found that nearly all of the total nitrogen in the dissolved and colloidal form was free ammonia. It is thus concluded that the amounts of free ammonia reported in the McCarthy tests are not appreciably in error.

The temperature of the mixtures during the experiments by McCarthy (not shown in Tables 4 to 9, inclusive) was from 22°C to 26°C. According to Morris, one of the possible reasons for high results in the determination of free residual chlorine in the presence of chloramines by either the amperometric titration of the orthotolidine test is that these tests do not sharply differentiate between free and combined residual chlorine when the test is conducted above about 18°C. Morris states that accuracy may be improved by cooling the test solutions to about 10°C before conducting the test for free residual chlorine. This was done by McCarthy who found better differentiation and much better reproducibility at around 10°C. The 1960 edition of "Standard



Methods<sup>2</sup> calls for chilling to as near 1°C as possible, and states that the temperature of the sample should never exceed 20°C when it is mixed with the orthotolidine.

In the tests reported in Tables 4 to 9 it will be noted that both the total and free residuals decrease over along periods of time indicating that the reactions were quite slow. McCarthy has made additional tests with 0.2% sewage, 0.6% sewage, 1.2% sewage and 1.3% sewage. A free chlorine residual was found after various intervals of time in decreasing amounts up to 23 hr after dosing. Free ammonia was also found in the chlorinated mixtures, both immediately after dosing and at the end of the test period, the amount of free ammonia having decreased materially after several hours.

On July 13, 1961, Culver applied a 15 ppm dose of chlorine to a 50% mixture of centrifuged Lawrence sewage with ammonia-free distilled water. The samples were cooled to 10°C to make the tests for residual chlorine. A free chlorine residual of 0.17 ppm was found after two minutes by the OTA test. The mixture contained about 35 ppm of free ammonia nitrogen. The temperature of the mixture was about 28°C and the pH about 7.1. The ratio of chlorine to free ammonia nitrogen was 1 to 2.3. No free chlorine residual was found after 50 min and 16 hr by the OTA tests or after 50 min by amperometric titration. The total oxidizing chlorine by the iodometric method was 8 ppm after 2 min, 6.7 ppm after 50 min, and 3.1 ppm after 16 hr. The results were about as might be expected from the known reactions of chlorine and ammonia in water, except for the small amount of free chlorine after 2 min.

On July 14, 1961, Culver applied a chlorine dose of 31 ppm to undiluted centrifuged Lawrence sewage containing about 42 ppm of free ammonia nitrogen. The temperature was 25°C and the pH was 7.45. The ratio of chlorine to free-ammonia nitrogen was 1 to 1.35. The samples were cooled to 10°C to make the tests for residual chlorine. In this case, the OTA tests showed a free chlorine residual of 1.0 ppm after 2 min, of 0.5 ppm after 15 min and of 0.18 ppm after 45 min. The total oxidizing chlorine residual by the iodometric method was 18.6 ppm after 2 min, 16.0 ppm after 25 min and 16.0 ppm after 45 min.

Both the OTA test and the amperometric test for free residual chlorine are based on the assumption that free residual chlorine reacts very rapidly with the reagent and combined chlorine reacts so slowly with the same reagent that the amount of combined chlorine taking part in the reaction is negligible by comparison with the free chlorine. In the OTA test the reagent is orthotolidine which is supposed to react almost instantaneously with the free chlorine. Within 5 sec the arsenite reagent is added to reduce the combined active chlorine without interference with the completed orthotolidine reaction. In the amperometric titration method, the sample is maintained between pH 6 and 7.5 by buffering if necessary, a range in which the combined chlorine is supposed not to react with the reagent. The sample is titrated with phenylarsenoxide solution to a null point indicated by the microammeter reading. To the sample remaining from the free chlorine titration, potassium iodide solution is added to convert the combined active chlorine to chloride releasing an equivalent amount of iodine. This equivalent iodine is then titrated with phenylarsenoxide solution to determine the combined available chlorine.

In samples containing a high proportion of combined available chlorine, more free available chlorine may be indicated by the OTA method than is actually present. Accordingly, when there is no free available chlorine and a

high combined available chlorine, part of the combined available chlorine will appear as free available chlorine. Similarly with the amperometric titration method if the sample contains a high concentration of combined chlorine, an apparent increase in the free chlorine reading may occur. It therefore follows that if the sample contains only combined chlorine in high concentrations, part of the combined chlorine may appear in the titration as free chlorine.

It should be evident from the preceding comments that no free chlorine was present in the McCarthy experiments, Tables 4 to 9, inclusive. The method used to determine the "combined chlorine demand" did not in fact yield the combined chlorine demand because the apparent free chlorine residual of 0.05 ppm was in fact part of the combined chlorine. It is reasonable to suppose that the "combined chlorine demand" could have been estimated more reliably by multiplying the concentration of free ammonia nitrogen by a factor of 10. Tests could then have been made with chlorine doses bracketing the computed dose to determine the "break point." It is probable that with chlorine doses of this magnitude, 16 ppm to 62 ppm, the reactions between the chlorine and the ammonia would be much more rapid than actually occurred in the McCarthy tests.

TABLE 12.—FLOW AND SUSPENDED SOLIDS, BIRD AVENUE DISTRICT, BUFFALO, N. Y., AUGUST 19, 1936

Time (P.M.)	Flow, in mgd	Suspended Solids		Suspended Solids Multiple of D. W. Rate, in lb per day
		in ppm	in lb per day	
	Average D. W. Rate			
	23.4	186	36,200	1
5:30	25.3	208	44,000	1.2
6:30	120.6	964	970,000	26.9
7:30	104	1,220	1,060,000	29.3
8:30	56.7	696	330,000	9.1
9:30	37.6	364	114,000	3.2
10:30	28.6	172	41,000	1.1
Average Multiple				11.8

*Loss of Suspended Solids and BOD in Overflows.*—The 1938-1940 chlorine demand studies by Symons<sup>14</sup> et al show that, on the average, the chlorine demand of the combined sewage during runoff from rainstorms is about 11% greater than the average chlorine demand of the dry-weather flow. Since a large number of chlorine demand determinations were made both during dry weather and during 126 days when rain occurred, the average values should be quite reliable for Buffalo sewage. It is evident from these data that there must have been a somewhat similar increase in the concentration of organic matter picked up from the bottoms of the sewers, and that the overflows of mixed sewage and storm water contained this increased concentration of solids.

Studies by Riis-Carstensen<sup>15</sup> of gagings and analyses of Buffalo sewage in the Bird Avenue district during August, 1936, throw light on the suspended solids discharged in overflows. Table 12 shows data presented by Riis-

<sup>15</sup> "Improving the Efficiency of Existing Interceptors," by Erik Riis-Carstensen, *Sewage and Industrial Wastes*, Vol. 27, 1955, p. 1115.

Carstensen for a rainstorm amounting to 0.55 in which started at 5:30 PM and ended at 9:00 PM. The writer has rearranged the results in the interest of clarity. The rainfall intensity averaged about 0.16 in. per hr, which should produce an average dilution ratio of about 14 to 1 after the first wetting, based upon 0.01 in. per hr as the rainfall rate equivalent to the dry weather flow. The maximum gaged rate of flow, 120.6 mgd, corresponds with a dilution ratio of only 5.2. According to Riis-Carstensen, a series of overflows are located upstream of a bottle-neck and of the gaging point in the Bird Avenue trunk sewer; and a flow estimated at about 50% of the total was discharged through these overflows.

Table 12 indicates that the rate of flow of suspended solids by weight reached 29.3 times the dry weather rate and averaged 11.8 times the dry weather rate during runoff from the rainstorm. If 50% of the flow was lost through the upstream overflows, the average rate of flow of suspended solids was about 24 times the dry weather rate.

If it is assumed that the average concentration of suspended solids and of BOD in the mixed sewage and storm water increases during runoff by 11%, as shown by Simons et al for the chlorine demand, and the storm water runoff amounts to about 40% of the years' dry weather flow, the total weight of BOD or of suspended solids which will be discharged to the receiving stream in the overflows will be about 44.5% of the year's production in the dry weather flow ( $40\% \times 1.11 = 44.5\%$ ). This is an astonishingly high figure, but the order of magnitude may be verified by the Riis-Carstensen data.

Using the measured data for the Bird Avenue district in Buffalo during August, 1936, and assuming that overflows occur 3% of the time, the total weight of suspended solids in the overflows will be about 35% of the year's production of suspended solids in the dry-weather flow ( $11.8 \times 3\% = 35\%$ ). Correcting the flow to account for 50% loss through upstream overflows, the suspended solids discharged through overflows will be about 70% of the total year's production in the dry-weather flow.

The Buffalo data tend to indicate that the annual loss of suspended solids and BOD to the receiving waters in overflows of mixed sewage and storm water is about half the total weight reaching the sewage treatment plant through the interceptors. This means that about 33% of the year's production of suspended solids and BOD overflows without treatment even though only 2% to 3% of the sewage overflows. There is no reason to suppose that this condition is not general with all combined sewerage systems, with variations in magnitude, of course. This is a very important finding, the significance of which appears to have been overlooked by most of the sanitary engineering profession, including the writer.

The effect of these estimates on the performance of primary and complete treatment plants serving combined sewerage systems through interceptors collecting two to three times the average dry weather flow is extremely important. Assuming the loss of suspended solids and BOD through overflows is 50% of the dry weather flow of suspended solids and BOD throughout the year, a primary plant removing 60% of the suspended solids and 30% of the BOD in the dry weather flow is actually removing only 40% of the total suspended solids ( $\frac{100}{150} \times 60 = 40\%$ ) and only 20% of the BOD ( $\frac{100}{150} \times 30 = 20\%$ ). Similarly a complete treatment plant removing 90% of the BOD and suspended

solids in the dry weather flow is actually removing only 60% of the total produced ( $\frac{100}{150} \times 90 = 60\%$ ).

Measurements of the per capita production of BOD and suspended solids based on analyses at treatment plants serving combined sewerage systems should be corrected to reflect the losses of solids through overflows. Assuming again that the loss is 50% by weight of solids reaching the treatment plant, the total per capita production should be increased by 50% over the figures indicated by the analyses at the treatment plants. For example, a BOD determination of 0.16 lb per day per capita at the plant should be increased to 0.24 lb per day per capita total.

It is evident from the preceding comments that, where the water quality standards for the receiving waters require removal of suspended solids or BOD, failure to account for the suspended solids and BOD in the overflows from combined systems will produce gross errors in the removals expected. In such cases, provisions should be made for returning to the system after the storms subside the solids settled in chlorine contact chambers provided for the overflows.

*Errata.*—Table 5, 80-minute free chlorine residual should read 0.05 instead of 0.5. Table 5, total chlorine residuals should be multiplied by 2 to read downward, 2.50, 2.06, 2.06, 1.66 and 1.66. Table 8, free  $\text{NH}_3\text{-N}$  for undiluted sewage should read 62 instead of 6.2.



RADIOLOGICAL CURRICULA FOR SANITARY ENGINEERS<sup>a</sup>

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Discussion by Gilbert H. Dunstan

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GILBERT H. DUNSTAN,<sup>3</sup> F. ASCE.—The authors have clearly stated the desirability of providing a course concerning the problems of radiation in sanitary engineering for every student studying for an advanced degree in sanitary engineering. They have suggested that this should provide approximately six semester-hours of credit, which may be difficult to work into a normal graduate program leading to a master's degree without omitting other important courses. To a large extent, the need for this much time will depend on the undergraduate preparation which the student has had, particularly in physics. If the student has had satisfactory work in modern physics, then it should be possible to provide a satisfactory course of 3 hr to 4 hr of credit. To some extent, this also depends on what radiological content has been worked into other courses, either undergraduate or graduate. A small amount of radiological work should be worked into such courses as water supply and waste water disposal, and into accompanying laboratory work, as the authors suggest. The writer fully concurs that this is a minimum coverage of the radiological field needed by future sanitary engineers regardless of specialty.

For those who will work directly in the radiological field, it is necessary, as the authors state, to provide additional course work in radiation subjects, because such a person should understand the sources of radiation exposure, the effects of radiation on man and animals, the means of protecting against radiation, and the problems of handling radioactive wastes. To do all this and still keep a solid foundation of other sanitary engineering and related courses is somewhat difficult, but not insurmountable, at the master's degree level. However, as emphasized, such a person is hardly to be considered a radiation specialist. This would require work at the doctor's degree level.

A person who has had considerable experience or training in sanitary engineering might be qualified to concentrate on radiological courses to a somewhat greater extent than that just indicated, by omitting certain sanitary engineering courses. He might then qualify for a type of radiation health specialist training which would prepare him for more specialized work in the radiological field than is normally required of sanitary engineers. He should not, however, be allowed to take such highly specialized training without an adequate background in sanitary engineering.

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<sup>a</sup> March 1961, by John E. Kiker, Jr. and F. Wellington Gilcreas (Proc. Paper 2767).

<sup>3</sup> Prof. of San. Engrg., Washington State Univ., Pullman, Wash.





# PROCEEDINGS PAPERS

The technical papers published in the past year are identified by number below. Technical-division sponsorship is indicated by an abbreviation at the end of each Paper Number, the symbols referring to: Air Transport (AT), City Planning (CP), Construction (CO), Engineering Mechanics (EM), Highway (HW), Hydraulics (HY), Irrigation and Drainage (IR), Pipeline (PL), Power (PO), Sanitary Engineering (SA), Soil Mechanics and Foundations (SM), Structural (ST), Surveying and Mapping (SU), and Waterways and Harbors (WW), divisions. Papers sponsored by the Department of Conditions of Practice are identified by the symbols (PP). For titles and order coupons, refer to the appropriate issue of "Civil Engineering." Beginning with Volume 82 (January 1956) papers were published in Journals of the various Technical Divisions. To locate papers in the Journals, the symbols after the paper number are followed by a numeral designating the issue of a particular Journal in which the paper appeared. For example, Paper 2703 is identified as 2703(ST1) which indicates that the paper is contained in the first issue of the Journal of the Structural Division during 1961.

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SEPTEMBER: 2588(IR3), 2589(IR3), 2590(WW3), 2591(IR3), 2592(HW3), 2593(IR3), 2594(IR3), 2595(IR3), 2596(HW3), 2597(WW3), 2598(IR3), 2599(WW3), 2600(WW3), 2601(WW3), 2602(WW3), 2603(WW3), 2604(HW3), 2605(SA5), 2606(WW3), 2607(SA5), 2608(ST9), 2609(SA5)<sup>c</sup>, 2610(IR3), 2611(WW3)<sup>c</sup>, 2612(ST9)<sup>c</sup>, 2613(IR3)<sup>c</sup>, 2614(HW3)<sup>c</sup>.  
 OCTOBER: 2615(EM5), 2616(EM5), 2617(ST10), 2618(SM5), 2619(EM5), 2620(EM5), 2621(ST10), 2622(EM5), 2623(SM5), 2624(EM5), 2625(SM5), 2626(SM5), 2627(EM5), 2628(EM5), 2629(ST10), 2630(ST10), 2631(PO5)<sup>c</sup>, 2632(EM5)<sup>c</sup>, 2633(ST10), 2634(ST10), 2635(ST10)<sup>c</sup>, 2636(SM5)<sup>c</sup>.  
 NOVEMBER: 2637(ST11), 2638(ST11), 2639(CO3), 2640(ST11), 2641(SA6), 2642(WW4), 2643(ST11), 2644(HY9), 2645(ST11), 2646(HY9), 2647(WW4), 2648(WW4), 2649(WW4), 2650(ST11), 2651(CO3), 2652(HY9), 2653(HY9), 2654(ST11), 2655(HY9), 2656(HY9), 2657(SA6), 2658(WW4), 2659(WW4)<sup>c</sup>, 2660(SA6), 2661(CO3), 2662(CO3), 2663(SA6), 2664(CO3)<sup>c</sup>, 2665(HY9)<sup>c</sup>, 2666(SA6)<sup>c</sup>, 2667(ST11)<sup>c</sup>.  
 DECEMBER: 2668(ST12), 2669(IR4), 2670(SM6), 2671(IR4), 2672(IR4), 2673(IR4), 2674(ST12), 2675(EM6), 2676(IR4), 2677(HW4), 2678(ST12), 2679(EM6), 2680(ST12), 2681(SM6), 2682(IR4), 2683(SM6), 2684(SM6), 2685(IR4), 2686(EM6), 2687(EM6), 2688(EM6), 2689(EM6), 2690(EM6), 2691(EM6)<sup>c</sup>, 2692(ST12), 2693(ST12), 2694(HW4)<sup>c</sup>, 2695(IR4)<sup>c</sup>, 2696(SM6)<sup>c</sup>, 2697(ST12)<sup>c</sup>.

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JANUARY: 2698(PP1), 2699(PP1), 2700(HY1), 2701(SA1), 2702(SU1), 2703(ST1), 2704(ST1), 2705(SU1), 2706(HY1), 2707(HY1), 2708(HY1), 2709(PO1), 2710(HY1), 2711(HY1), 2712(ST1), 2713(HY1), 2714(PO1), 2715(ST1), 2716(HY1), 2717(SA1), 2718(SA1), 2719(SU1)<sup>c</sup>, 2720(SA1)<sup>c</sup>, 2721(ST1), 2722(PP1)<sup>c</sup>, 2723(PO1)<sup>c</sup>, 2724(HY1)<sup>c</sup>, 2725(ST1)<sup>c</sup>.  
 FEBRUARY: 2726(WW1), 2727(EM1), 2728(EM1), 2729(WW1), 2730(WW1), 2731(EM1), 2732(SM1), 2733(WW1), 2734(SM1), 2735(EM1), 2736(EM1), 2737(PL1), 2738(PL1), 2739(PL1), 2740(PL1), 2741(EM1), 2742(ST2), 2743(EM1), 2744(WW1), 2745(EM1), 2746(SM1), 2747(WW1), 2748(EM1), 2749(WW1), 2750(WW1)<sup>c</sup>, 2751(EM1)<sup>c</sup>, 2752(SM1)<sup>c</sup>, 2753(PL1)<sup>c</sup>, 2754(ST2)<sup>c</sup>, 2755(PL1).  
 MARCH: 2756(HY2), 2757(IR1), 2758(AT1), 2759(CO1), 2760(HY2), 2761(IR1), 2762(IR1), 2763(HY2), 2764(ST3), 2765(HY2), 2766(HW1), 2767(SA2), 2768(CO1), 2769(IR1), 2770(HY2), 2771(SA2), 2772(HY2), 2773(CO1), 2774(AT1), 2775(IR1), 2776(HY2), 2777(HY2), 2778(SA2), 2779(ST3), 2780(HY2), 2781(HY2)<sup>c</sup>, 2782(HW1)<sup>c</sup>, 2783(SA2)<sup>c</sup>, 2784(CO1), 2785(CO1)<sup>c</sup>, 2786(IR1)<sup>c</sup>, 2787(ST3)<sup>c</sup>, 2788(AT1)<sup>c</sup>, 2789(HW1).  
 APRIL: 2790(EM2), 2791(SM2), 2792(SM2), 2793(SM2), 2794(SM2), 2795(SM2), 2796(SM2), 2797(SM2), 2798(EM2), 2799(EM2), 2800(EM2), 2801(EM2), 2802(ST4), 2803(EM2)<sup>c</sup>, 2804(SM2)<sup>c</sup>, 2805(ST4)<sup>c</sup>.  
 MAY: 2806(SA3), 2807(WW2), 2808(HY3), 2809(WW2), 2810(HY3), 2811(WW2), 2812(HY3), 2813(WW2), 2814(HY3), 2815(WW2), 2816(HY3), 2817(HY3), 2818(SA3), 2819(WW2), 2820(SA3), 2821(WW2), 2822(WW2)<sup>c</sup>, 2823(HY3), 2824(SA3), 2825(HY3), 2826(SA3)<sup>c</sup>, 2827(HY3)<sup>c</sup>.  
 JUNE: 2828(SM3), 2829(EM3), 2830(EM3), 2831(IR2), 2832(SM3), 2833(HW2), 2834(IR2), 2835(EM3), 2836(IR2), 2837(IR2), 2838(SM3), 2839(SM3)<sup>c</sup>, 2840(IR2)<sup>c</sup>, 2841(HW2)<sup>c</sup>, 2842(EM3)<sup>c</sup>, 2843(ST5), 2844(ST5), 2845(ST5), 2846(ST5)<sup>c</sup>.  
 JULY: 2847(PO2), 2848(SU2), 2849(HY4), 2850(PO2), 2851(HY4), 2852(PO2), 2853(SU2), 2854(HY4), 2855(PO2), 2856(PO2), 2857(PO2), 2858(SA4), 2859(SU2), 2860(SA4), 2861(PO2), 2862(SA4), 2863(HY4), 2864(HY4), 2865(HY4), 2866(HY4), 2867(HY4), 2868(PO2)<sup>c</sup>, 2869(SA4)<sup>c</sup>, 2870(SU2)<sup>c</sup>, 2871(HY4), 2872(HY4)<sup>c</sup>, 2873(SU2), 2874(SA4).  
 AUGUST: 2875(WW3), 2876(WW3), 2877(WW3), 2878(SM4), 2879(ST6), 2880(EM4), 2881(SM4), 2882(EM4), 2883(WW3), 2884(EM4), 2885(SM4), 2886(WW3), 2887(EM4), 2888(WW3), 2889(AT2), 2890(AT2), 2891(AT2), 2892(AT2), 2893(AT2), 2894(AT2), 2895(AT2), 2896(AT2), 2897(AT2), 2898(AT2), 2899(AT2), 2900(AT2), 2901(AT2), 2902(SM4), 2903(ST6), 2904(ST6), 2905(SM4), 2906(ST6), 2907(EM4), 2908(ST6), 2909(EM4), 2910(ST6), 2911(EM4), 2912(SM4), 2913(ST6), 2914(WW3)<sup>c</sup>, 2915(ST6)<sup>c</sup>, 2916(EM4)<sup>c</sup>, 2917(SM4)<sup>c</sup>.  
 SEPTEMBER: 2918(SA5)<sup>c</sup>, 2919(HW3)<sup>c</sup>, 2920(HY5)<sup>c</sup>, 2921(SA5), 2922(PL2), 2923(IR3), 2924(HY5), 2925(HY5), 2926(CP1), 2927(HY5), 2928(IR3), 2929(IR3), 2930(HY5), 2931(CP1), 2932(PL2), 2933(HY5), 2934(HY5), 2935(HY5), 2936(HY5), 2937(HW3), 2938(CP1), 2939(PL2), 2940(SA5), 2941(SA5), 2942(SA5), 2943(HY5), 2944(PL2)<sup>c</sup>, 2945(CP1)<sup>c</sup>, 2946(IR3), 2947(HW3), 2948(IR3), 2949(IR3)<sup>c</sup>.

c. Discussion of several papers, grouped by divisions.

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